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PATENT ABSTRACTS OF JAPAN

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(54) MANUFACTURE OF RARE-EARTH BOND MAGNET

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a rare-earth bond magnet having a low hole rate, high dimensional precision, excellent moldability and excellent magnetic characteristics.

SOLUTION: First, rare-earth magnetic powder, binder resin composed of thermoplastic resin and antioxidant are mixed with predetermined ratios and the mixture is kneaded at a temperature higher than the heat deformation temperature. Then the kneaded mixture is granulated or rectified to obtain grains. The grains are subjected to the compression molding at a 1st temperature at which the thermoplastic resin is softened or melt and then cooled to a 2nd temperature which is at least lower than the 1st temperature while the pressure is applied and the thermoplastic resin is solidified to obtain a rare-earth bond magnet. It is recommended that the 2nd temperature is lower than the melting point or the heat deformation temperature of the thermoplastic resin.

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CLAIMS

[Claim(s)]

[Claim 1] A manufacture method of a rare earth bond magnet which comes to join together rare earth magnet powder characterized by providing the following with joint resin which consists of thermoplastics A process which mixes and kneads said rare earth magnet powder and said joint resin, and manufactures a kneading object They are a granulation or the process which carries out a particle size regulation and which is used as a granular object about said kneading object. A process which carries out pressing at the 1st temperature from which said joint resin will be in softening or a melting condition using said granular object A process cooled in the state of application of pressure to the 2nd temperature which is said under 1st temperature at least

[Claim 2] Said kneading is the manufacture method of a rare earth bond magnet according to claim 1 which is the temperature more than heat deflection temperature of said joint resin, and is performed so that a front face of said rare earth magnet powder may be in the condition of having been covered with melting or a softened joint resinous principle.

[Claim 3] A manufacture method of a rare earth bond magnet according to claim 1 or 2 that a content of said rare earth magnet powder in said kneading object is 90 – 99wt%.

[Claim 4] A manufacture method of a rare earth bond magnet according to claim 1 to 3 which contains an anti-oxidant in said kneading object.

[Claim 5] A manufacture method of a rare earth bond magnet according to claim 4 that a content of said anti-oxidant in said kneading object is 0.1 – 2wt%.

[Claim 6] Said granulation or particle size regulation is the manufacture method of a rare earth bond magnet according to claim 1 to 5 performed by grinding.

[Claim 7] mean particle diameter of said granular object — 10 micrometers – a manufacture method of a rare earth bond magnet according to claim 1 to 6 which is 2mm.

[Claim 8] Said pressing is the manufacture method of a rare earth bond magnet according to claim 1 to 7 which is compression molding.

[Claim 9] Said 2nd temperature is the manufacture method of a rare earth bond magnet according to claim 1 to 8 which is the melting point of said joint resin.

[Claim 10] Said 2nd temperature is the manufacture method of a rare earth bond magnet according to claim 1 to 8 which is the heat deflection temperature of said joint resin.

[Claim 11] A manufacture method of a rare earth bond magnet according to claim 1 to 10 that a difference of said 1st temperature and said 2nd temperature is 20 degrees C or more.

[Claim 12] Cooling in said application-of-pressure condition is the manufacture method of a rare earth bond magnet according to claim 1 to 11 performed continuously, without canceling application of pressure in the case of said pressing.

[Claim 13] A manufacture method of a rare earth bond magnet according to claim 1 to 12 that a pressure at the time of cooling in said application-of-pressure condition is less than [an EQC or it] to compacting pressure at the time of said pressing.

[Claim 14] A pressure at the time of cooling in said application-of-pressure condition is the manufacture method of a rare earth bond magnet according to claim 1 to 13 currently uniformly held to the melting point of said joint resin at least.

[Claim 15] A pressure at the time of cooling in said application-of-pressure condition is the manufacture method of a rare earth bond magnet according to claim 1 to 13 currently uniformly held to temperature between said 1st temperature and 2nd temperature at least.

[Claim 16] A cooling rate at the time of cooling in said application-of-pressure condition is the manufacture method of a rare earth bond magnet according to claim 1 to 15 which is 0.5–100 degrees C/second.

[Claim 17] Compacting pressure at the time of said pressing is 2 60 kgf/mm. A manufacture method of a rare earth bond magnet according to claim 1 to 16 which is the following.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] This invention relates to the manufacture method of a rare earth bond magnet, and the constituent for rare earth bond magnets.

[0002]

[Description of the Prior Art] Pressing of the rare earth bond magnet is carried out to the magnet configuration of a request of the mixture (compound) of rare earth magnet powder and joint resin (organic binder), and it is manufactured. It divides roughly into the shaping method by this pressing, and there are compression forming, an injection-molding method, and an extrusion method in it.

[0003] Compression forming is the method of being filled up with said compound into press metal mold, pressing this at predetermined temperature and acquiring a Plastic solid, making harden it after that, when joint resin is thermosetting resin, and using as a magnet. Compared with other methods, the amount of resin of this method in the magnet with which the amount of joint resin was obtained since it was able to fabricate at least decreases, and it is advantageous for improvement in magnetic properties.

[0004] An extrusion method is the method of carrying out cooling solidification, while extruding said compound by which heating melting was carried out from the metal mold of an extruding press machine, and cutting to the desired length and using as a magnet. Although there is an advantage that the flexibility to a magnetic configuration is large and the magnet of thin meat and a long picture can also be manufacture easily , by this method , in order to secure the fluidity of the melt at the time of shaping , there is a defect that there are many amounts of resin in the magnet which needed to make [many] the addition of joint resin compared with it of compression forming , therefore was obtained , and there is an inclination for magnetic properties to fall .

[0005] An injection-molding method is the method of carrying out heating fusion of said compound, pouring in this melt into metal mold, where sufficient fluidity is given, and fabricating in a predetermined magnet configuration. By this method, the flexibility to a magnetic configuration has the advantage that it is still larger compared with an extrusion method, and a variant magnet can also be manufactured especially easily. However, since level with the fluidity of the melt at the time of shaping higher than said extrusion method is required, the addition of joint resin has the defect that there are many amounts of resin in the magnet which needed to make [more / still] it compared with it of an extrusion method, therefore was obtained, and there is an inclination for magnetic properties to fall.

[0006]

[Problem(s) to be Solved by the Invention] By the way, compression molding which can fabricate a magnet with the magnetic highest engine performance among the above all directions methods has a defect [like / next].

[0007] Although the density of a Plastic solid is high, in order that the rare earth bond magnet manufactured by the 1st may show the inclination for a void content to become high, its mechanical strength is weak and it is inferior to corrosion resistance in it. Therefore, it sets especially to compression forming and is compacting pressure 70 kgf(s)/mm² High-pressure molding made into the above and high voltage was utilized, and it was coped with by performing coating processing for corrosion prevention etc. after shaping. However, the burden of high-pressure molding to that of a making machine is large, and when

performing coating processing for corrosion prevention, the process for it is added and it causes lowering of the productivity by complication of a manufacturing process, and lifting of a manufacturing cost.

[0008] Since resin is in the condition of not hardening, change of the physical properties by hardening of resin and change of the physical properties by water absorption arise, and, as for the compound which used [2nd] thermosetting resin, the moldability of a compound changes with time. By this, even when it fabricates on the same conditions, the size and density of a Plastic solid change, and fabricating to stability becomes difficult. Moreover, when thermosetting resin is used, in order a curing (hardening) process is needed, and change of the size by the reaction of the resin at the time of curing arises it not only leads to an increment and cost rise of a process by this, but and to secure an aim size, amendment of a metal mold size is needed and size reservation is not easy.

[0009] Moreover, in the case of the conventional compression molding, both solid thing and liquefied thing are used at a room temperature as thermosetting resin. Among these, when the former solid-state resin is used, although ***** is comparatively good, its moldability is bad, and the inclination for a void content to become high more is shown. Moreover, the dispersibility of resin and magnet powder is bad, consequently a mechanical strength falls. On the other hand, although it is possible to acquire the Plastic solid of high density when the latter liquefied resin is used, the physical properties of resin change sensitively under the effect by the environment at the time of shaping (temperature, humidity), and the restoration nature to metal mold falls.

[0010] Therefore, variation arises to a magnetic aim size, namely, dimensional accuracy is bad and lacks in the stability of shaping. Especially, in the case of a small magnet, this defect becomes remarkable. Thus, since the variation in a size is large, in order to secure the aim size of the last magnet product, after fabricating more greatly than an aim size, fabricating, such as a cut and polishing, needs to adjust a size. Since the increment in a process is caused and a poor material is generated by processing by this, productivity falls and a manufacturing cost increases. Moreover, in order to cancel such a defect, a special device must be given to the structure and the forming cycle of a making machine, consumption of a making machine is also remarkable and the cycle time of shaping also becomes long.

[0011] Furthermore, it is one of the causes that the 1st and 2nd above defects also have unsuitable manufacture method of a compound, manufacture conditions, temperature conditions at the time of shaping, cooling conditions after shaping, etc.

[0012] Therefore, the object of this invention is to offer the manufacture method of a rare earth bond magnet that the rare earth bond magnet of the low void content excellent in a moldability, magnetic properties, and dimensional stability can be manufactured easily.

[0013]

[Means for Solving the Problem] Such an object is attained by this invention of following the (1) – (17).

[0014] (1) A process which is the manufacture method of a rare earth bond magnet which comes to join rare earth magnet powder together with joint resin which consists of thermoplastics, mixes and kneads said rare earth magnet powder and said joint resin, and manufactures a kneading object, A process which carries out pressing of said kneading object at a granulation or a process which carries out a particle size regulation, and which is used as a granular object, and the 1st temperature from which said joint resin will be in softening or a melting condition using said granular object, A manufacture method of a rare earth bond magnet characterized by having a process cooled in the state of application of pressure to the 2nd temperature which is said under 1st temperature at least.

[0015] (2) Said kneading is the manufacture method of a rare earth bond magnet given in the above (1) which is the temperature more than heat deflection temperature of said joint resin, and is performed so that a front face of said rare earth magnet powder may be in the condition of having been covered with melting or a softened joint resinous principle.

[0016] (3) A manufacture method of a rare earth bond magnet given in the above 1 or 2 whose contents of said rare earth magnet powder in said kneading object are 90 – 99wt%.

[0017] (4) The above (1) which contains an anti-oxidant in said kneading object thru/or a manufacture method of a rare earth bond magnet given in either of (3).

[0018] (5) A manufacture method of a rare earth bond magnet given in the above (4) whose content of said anti-oxidant in said kneading object is 0.1 – 2wt%.

[0019] (6) Said granulation or particle size regulation is the manufacture method of a rare earth bond magnet the above (1) performed by grinding thru/or given in either of (5).

[0020] (7) mean particle diameter of said granular object — 10 micrometers – a manufacture method of a rare earth bond magnet the above (1) which is 2mm thru/or given in either of (6).

[0021] (8) Said pressing is the manufacture method of a rare earth bond magnet the above (1) which is compression molding thru/or given in either of (7).

[0022] (9) Said 2nd temperature is the manufacture method of a rare earth bond magnet the above (1) which is the melting point of said joint resin thru/or given in either of (8).

[0023] (10) Said 2nd temperature is the manufacture method of a rare earth bond magnet the above (1) which is the heat deflection temperature of said joint resin thru/or given in either of (8).

[0024] (11) The above (1) whose difference of said 1st temperature and said 2nd temperature is 20 degrees C or more thru/or a manufacture method of a rare earth bond magnet given in either of (10).

[0025] (12) Cooling in said application-of-pressure condition is the manufacture method of a rare earth bond magnet the above (1) performed continuously, without canceling application of pressure in the case of said pressing thru/or given in either of (11).

[0026] (13) The above (1) whose pressure at the time of cooling in said application-of-pressure condition is less than [an EQC or it] to compacting pressure at the time of said pressing thru/or a manufacture method of a rare earth bond magnet given in either of (12).

[0027] (14) A pressure at the time of cooling in said application-of-pressure condition is the manufacture method of a rare earth bond magnet the above (1) currently uniformly held to the melting point of said joint resin at least thru/or given in either of (13).

[0028] (15) A pressure at the time of cooling in said application-of-pressure condition is the manufacture method of a rare earth bond magnet the above (1) currently uniformly held to temperature between said 1st temperature and 2nd temperature at least thru/or given in either of (13).

[0029] (16) A cooling rate at the time of cooling in said application-of-pressure condition is the manufacture method of a rare earth bond magnet the above (1) which is 0.5–100 degrees C/second thru/or given in either of (15).

[0030] (17) Compacting pressure at the time of said pressing is 2 60 kgf/mm. The above (1) which is the following thru/or a manufacture method of a rare earth bond magnet given in either of (16).

[0031]

[Embodiment of the Invention] Hereafter, the manufacture method of the rare earth bond magnet of this invention is explained to details.

[0032] The manufacture method of the rare earth bond magnet of this invention mainly has the following processes.

[0033] Manufacture **** of the kneading object of the constituent for <1> rare-earth bond magnets and the constituent for rare earth bond magnets (only henceforth a "constituent") are adjusted. This constituent mainly consists of rare earth magnet powder and joint resin (binder). Moreover, an antioxidant is contained preferably and other additives are added if needed. It is mixed using mixers and agitators, such as a Henschel mixer, further, each of these constituents are kneaded so that it may mention later, and they obtain a kneading object.

[0034] Hereafter, each of these constituents are explained.

[0035] 1. What consists of an alloy containing rare earth elements and transition metals as rare earth magnet powder rare earth magnet powder is desirable, and following [1] – [4] is desirable especially.

[0036] [1] What makes a fundamental component the rare earth elements which are mainly concerned with Sm, and the transition metals which are mainly concerned with Co (henceforth a Sm-Co system alloy).

[0037] [2] What makes a fundamental component the transition metals which are mainly concerned with R (at least one sort however, among the rare earth elements with which R contains Y), and Fe, and B (henceforth a R-Fe-B system alloy).

[0038] [3] What makes a fundamental component the rare earth elements which are mainly concerned with Sm, the transition metals which are mainly concerned with Fe, and the element between grids which is mainly concerned with N (henceforth a Sm-Fe-N system alloy).

[0039] [4] What makes a fundamental component transition metals, such as R (at least one sort however, among the rare earth elements with which R contains Y), and Fe, and has a magnetic phase on nano meter level (henceforth a "nano crystal magnet").

[0040] [5] The above [1] What mixed at least two sorts in the thing of a presentation of - [4]. In this case, it can have the advantage of each magnet powder to mix simulataneously, and more excellent magnetic properties can be acquired easily.

[0041] As a typical thing of a Sm-Co system alloy, SmCo₅ and Sm₂ TM₁₇ (however, TM, transition metals) are mentioned.

[0042] As a typical thing of a R-Fe-B system alloy, a Nd-Fe-B system alloy, a Pr-Fe-B system alloy, a Nd-Pr-Fe-B system alloy, a Ce-Nd-Fe-B system alloy, a Ce-Pr-Nd-Fe-B system alloy, the thing that replaced a part of Fe in these with other transition metals, such as Co and nickel, are mentioned.

[0043] It is Sm₂ Fe₁₇N₃ which nitrided and produced Sm₂ Fe₁₇ alloy as a typical thing of a Sm-Fe-N system alloy. It is mentioned.

[0044] as said rare earth elements in magnet powder, Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and a misch metal mention -- having -- these -- one sort -- or two or more sorts can be included. moreover, Fe, Co, nickel, etc. mention as said transition metals -- having -- these -- one sort -- or two or more sorts can be included. Moreover, in order to raise magnetic properties, in magnet powder, B, aluminum, Mo, Cu, Ga, Si, Ti, Ta, Zr, Hf, Ag, Zn, etc. can also be contained if needed.

[0045] Moreover, especially the mean particle diameter of magnet powder is 0.5-100 micrometers, although not limited. A degree is desirable and it is 1-50 micrometers. A degree is more desirable. in addition, the particle size of magnet powder etc. -- for example, F.S.S.S. (Fischer Sub-Sieve Sizer) -- it can measure by law.

[0046] Moreover, even when the particle size distribution of magnet powder is uniform, even if it distributes to some extent (there is variation), it is good, but latter one is desirable in order to obtain the good moldability at the time of shaping by a small amount of joint resin which is mentioned later. Thereby, the void content of the obtained bond magnet can also be reduced more. In addition, in the above [5], the mean particle diameter may differ for every presentation of the magnet powder to mix.

[0047] It may be the quenching thin band manufacturing installation used for especially the manufacture method of magnet powder not being limited, for example, producing an alloy ingot by dissolution and casting, grinding this alloy ingot to a moderate grain size, and manufacturing a ***** (classifying further) thing and an amorphous alloy, and any are [a ribbon-like quenching flake (detailed polycrystals gather) may be manufactured, this flake (thin band) may be ground to a moderate grain size, and] sufficient as a ***** (classifying further) thing etc.

[0048] It is desirable that it is about 90-99wt%, as for the content in the inside of the kneading object of the above magnet powder, it is more desirable that it is about 93-99wt%, and it is more desirable that it is about 95-99wt%. If improvement in magnetic properties (especially magnetic energy product) cannot be aimed at if there are too few contents of magnet powder, and there are too many contents of magnet powder, the content of joint resin will decrease relatively and a moldability will fall.

[0049] 2. Joint Resin (Binder)

Thermoplastics is used as joint resin (binder). Although it is advantageous compared with the case where thermosetting resin is used when obtaining the magnet of a low void content when thermoplastics is used as joint resin, in this invention, the temperature conditions at the time of shaping mentioned later, cooling conditions, and a conjointly more low void content are realizable.

[0050] As thermoplastics, for example A polyamide (example: nylon 6, Nylon 46, Nylon 66, Nylon 610, Nylon 612, Nylon 11, Nylon 12, Nylon 612, nylon 6 -66), Liquid crystal polymers, such as thermoplastic polyimide and aromatic polyester system resin, Polyphenylene oxide, polyphenylene sulfide, polyethylene, Polyolefines, such as polypropylene and an ethylene-vinylacetate copolymer, The copolymer which is mainly concerned

with these, a blend object, a polymer alloy, etc. are mentioned, and denaturation polyolefine, a polyether, polyacetal, etc. can mix and use 1 of sorts of these, and two sorts or more.

[0051] among these — also coming out — that which is mainly concerned with polyolefine is desirable in respect of that which is excellent in a moldability, and is mainly concerned with a liquid crystal polymer and polyphenylene sulfide from the point of a polyamide or its copolymer, and heat-resistant improvement since the mechanical strength is strong, the ease of shaping, or low cost. Moreover, these thermoplastics is excellent also in kneading nature with magnet powder.

[0052] It is desirable that the melting point is a thing 120 degrees C or more, as for the thermoplastics used, it is more desirable that it is what is 122 degrees C – 400 degrees C, and it is still more desirable that it is what is 125 degrees C – 350 degrees C. The thermal resistance of a magnet Plastic solid falls that the melting point is the thing of under said lower limit, and it becomes difficult to secure sufficient temperature characteristic (magnetic or mechanical). Moreover, the temperature at the time of shaping rises that it is that to which the melting point exceeds said upper limit, and it becomes easy to produce oxidation of magnet powder etc.

[0053] Moreover, in order to raise a moldability more, as for the average molecular weight (polymerization degree) of the thermoplastics used, it is desirable that it is 10000 to about 60000, and it is more desirable that it is 10000 to about 35000.

[0054] It is desirable that it is about 1–10wt%, as for the content in the inside of the kneading object of the above joint resin, it is desirable that it is about 1–8wt%, and it is still more desirable that it is about 1–5wt%. When there are too many contents of joint resin, the inclination to be unable to aim at improvement in magnetic properties (especially the maximum magnetic energy product), and for dimensional accuracy to fall is shown. Moreover, if there are too few contents of joint resin, a moldability will fall.

[0055] 3. An antioxidant antioxidant is an additive added in this constituent, in order to prevent the deterioration (generated when the metal component of rare earth magnet powder works as a catalyst) by the oxidation degradation of rare earth magnet powder, or oxidation of joint resin in the case of manufacture of a kneading object etc. Addition of this antioxidant prevented oxidation of rare earth magnet powder, and it has played the role important at the time of kneading of the constituent for rare earth bond magnets, when contributing to improvement in the thermal stability at the time of shaping and securing a good moldability in the small amount of joint resin while it contributes to aiming at improvement in magnetic magnetic properties.

[0056] Since this antioxidant volatilizes or deteriorates in the time of kneading and shaping to a magnet etc., after that part has remained, it exists in the manufactured rare earth bond magnet.

[0057] The chelating agent which what kind of thing is sufficient as as an antioxidant as long as it can prevent or control oxidation of rare earth magnet powder etc., for example, generates a chelate compound to metal ions, such as an amine system compound, an amino acid system compound, nitro carboxylic acids, a hydrazine compound, a cyanide compound, and a sulfide, especially Fe component is used suitably. In addition, it cannot be overemphasized about the class of antioxidant, and a presentation that it is not limited to these things.

[0058] When adding such an antioxidant, as for the content of the antioxidant in a kneading object, considering as about 0.1–2wt% is desirable, and considering as about 0.5–1.5wt% is more desirable. In this case, as for the content of an antioxidant, it is desirable that it is about 2 – 150% to the content of joint resin, and it is more desirable that it is about 30 – 100%.

[0059] In addition, it cannot be overemphasized that the addition of an antioxidant may be below the lower limit of said range, and you may be additive-free in this invention.

[0060] The addition of said joint resin and antioxidant is determined with careful attention to the following.

[0061] That is, when there is little joint resin, the amount of magnet powder increases relatively and the viscosity of the kneading object in the case of kneading becomes high, and kneading torque increases and it becomes the inclination for oxidation of resin to be promoted by pyrexia. If there are few amounts of antioxidants at this time, it will become impossible to fully control oxidation of resin, viscosity lifting of a kneading object (resin melt) will arise, kneading nature and a moldability will fall, and the magnet which was

excellent in dimensional stability with the low void content and the high mechanical strength will not be obtained. Moreover, when there are many amounts of antioxidants, the amount of resin decreases relatively and the inclination for the mechanical strength of a Plastic solid to fall is shown.

[0062] On the other hand, when there is much joint resin, the amount of magnet powder decreases relatively, the effect to the resin of magnet powder falls, and oxidation of resin stops being able to happen easily. Therefore, an antioxidant becomes possible [controlling oxidation of resin at least].

[0063] Thus, if there are comparatively many contents of joint resin, the content of an antioxidant can be lessened, and if reverse has few contents of joint resin, it is necessary to make [many] the content of an antioxidant.

[0064] Therefore, as for the sum total content of the joint resin in a kneading object, and an antioxidant, it is desirable that it is 1.0 – 8.0wt%, and it is more desirable that it is 2.0 – 6.0wt%. By considering as such a range, it contributes to improvement in antioxidizing, such as a moldability at the time of shaping, the ease of shaping, and magnet powder, and the magnet of a low void content, a high mechanical strength, and high magnetic properties is obtained.

[0065] 4. In other additives and a kneading object, various additives, such as a plasticizer (for example, fatty-acid salts, such as zinc stearate, a fatty acid), lubricant (for example, various inorganic lubricant, such as silicone oil, various waxes, a fatty acid, an alumina, a silica, and a titania), and other shaping assistants, may be added if needed.

[0066] Since addition of a plasticizer raises the fluidity at the time of shaping, it makes it possible to be able to acquire the same property with the addition of fewer joint resin, and to press with lower moulding pressure. The same is said of addition of lubricant. As for the addition of a plasticizer, it is desirable that it is about 0.01–0.2wt%, and, as for the addition of lubricant, it is desirable that it is about 0.05–0.5wt%.

[0067] Other additives are mixed with an antioxidant if needed as preferably as the above rare earth magnet powder and joint resin, it kneads further, and a kneading object is manufactured.

[0068] Mixing is performed using mixers and agitators, such as a Henschel mixer.

[0069] Kneading is performed using kneading machines, such as for example, a biaxial extrusion kneading machine, a roll type kneading machine, and a kneader.

[0070] This kneading is preferably performed at the temperature more than the heat deflection temperature (it measures by the method by ASTM D648) of the joint resin to be used, and the temperature more than the melting point of the joint resin used more preferably.

[0071] For example, when a polyamide (heat deflection temperature of 145 degrees C, melting point of 178 degrees C) is used as joint resin, a desirable kneading temperature is about 150–280 degrees C. Moreover, although mixing time changes with the class of joint resin, and terms and conditions, such as kneading temperature, it is usually made into about 5 – 40 minutes.

[0072] Moreover, this kneading is fully performed so that the front face of rare earth magnet powder may be in the condition of having been covered with melting or the softened joint resinous principle. Although the mixing time for acquiring such a condition changes with terms and conditions, such as a class of joint resin, and a kneading machine to be used, kneading temperature, when it kneads at said kneading temperature, it is desirable to usually consider as about 5 – 90 minutes, and it is more desirable to consider as about 5 – 60 minutes.

[0073] Since it is kneaded after the viscosity of joint resin has fallen while being able to knead to homogeneity more in a short time compared with the case where the effectiveness of kneading improves and it kneads in ordinary temperature by kneading on such conditions, it will be in the condition that joint resin covered the perimeter of rare earth magnet powder to homogeneity, and it will contribute to reduction of the void content in the magnet with which the void content in a kneading object was decreased namely, manufactured.

[0074] in addition, the case where n kinds of thermoplastics is mixed and used as joint resin — the above “heat deflection temperature (or melting point) of the joint resin to be used” — for example, it is convertible as follows.

[0075] the amount of each thermoplastics when making the sum total of thermoplastics into 1 weight

section -- respectively -- A1 and A2 ... An the weight section and the heat deflection temperature (or melting point) of each thermoplastics -- respectively -- T1 and T2 ... Tn ** -- the heat deflection temperature (or melting point) of the thermoplastics used when it carries out -- A1 T1+A2 T2+ ... An Tn It is expressed. In addition, in the following processes, these conversion are made the same, also when mixing and using n kinds of thermoplastics.

[0076] the kneading object manufactured with the manufacture above <1> of <2> granulation object -- a granulation -- or a particle size regulation is carried out and the granular object of a predetermined particle size is manufactured.

[0077] Although especially the method of a granulation or a particle size regulation is not limited, it is desirable to be made by grinding a kneading object. This grinding is performed using a ball mill, a vibration mill, a crusher, a jet mill, a pin mill, etc.

[0078] Moreover, it can also carry out, for example using a granulating machine like an extrusion type granulating machine, and can also carry out further combining the granulation by the granulating machine, and said grinding.

[0079] Moreover, adjustment of the particle size of a granular object can be performed by classifying using a sieve etc.

[0080] the mean particle diameter of a granular object -- 10 micrometers -- it is about 2mm -- desirable -- 20 micrometers -- it is about 2mm -- more -- desirable -- 50 micrometers -- it is still more desirable that it is about 2mm. Since it becomes difficult to adjust the fill to the metal mold of a granular object delicately and quantum nature is inferior when the size of the magnet with which the mean particle diameter of a granular object is fabricated especially by 2mm or more is small (i.e., when the size of the gap of shaping metal mold is small), improvement in the dimensional accuracy of a bond magnet cannot be aimed at. On the other hand, it is the mean particle diameter of 10 micrometers. When the following granular objects may be difficult to manufacture (granulation), or may require time and effort and its mean particle diameter is too small, the inclination for the void content of the obtained bond magnet to rise is shown.

[0081] Although such a granular object has a certain amount of variation in particle size, what has a uniform particle size is desirable. Thereby, the pack density to metal mold increases and a bond magnet with high dimensional accuracy is obtained with a low void content.

[0082] In addition, the granular object said here is distinguished from a pellet with a large particle size (massive object).

[0083] Pressing is performed using the granular object obtained with <3> pressing above <2>. Hereafter, typical compression molding is explained.

[0084] It is filled up with a granulation object in the metal mold (gap) of a compacting machine, and it is pressed all over a magnetic field or a non-magnetic field (5-20kOe and the direction of orientation have an orientation magnetic field good also for any of length, width, and a radial direction).

[0085] This compression molding is performed by blue heat forming. That is, it considers as a predetermined temperature (the 1st temperature) from which the thermoplastics [heat / shaping metal mold] (joint resin) using the material temperature at the time of shaping will be in softening or a melting condition.

[0086] This 1st temperature is made into the temperature more than the heat deflection temperature of the thermoplastics to be used. Furthermore, it is desirable to consider as the temperature more than the melting point of the thermoplastics to be used, it is more desirable to consider as a predetermined temperature of the range from the melting point (melting point +200) to ** degree, and it is still more desirable to consider as a predetermined temperature of the range from the melting point (melting point +130) to ** degree.

[0087] For example, when the thermoplastics to be used is a polyamide (melting point: 178 degrees C), especially desirable material temperature at the time of shaping (the 1st temperature) is made into about 180-300 degrees C.

[0088] By fabricating at such a temperature, the fluidity of the molding material within metal mold improves, and not to mention the thing of cylindrical and the letter of a block, the thing of cylindrical (the shape of a

ring) and the configuration which has thin-walled parts, such as plate-like and bow tabular, a small thing, and a long picture thing also have a high mechanical strength, and can mass-produce the thing of fitness and the stable configuration, and a size with a low void content.

[0089] The compacting pressure in compression molding is 2 60 kgf(s)/mm preferably. It is 2 two to 50 kgf/mm more preferably hereafter. It is 2 five to 40 kgf/mm preferably to a degree and a pan. It considers as a degree. Since it fabricates at the 1st temperature which was mentioned above, the bond magnet which has the advantage which was mentioned above also with such comparatively low compacting pressure can be fabricated in this invention (size enlargement).

[0090] A Plastic solid is cooled after <4> cooling pressing. This cooling is performed in the state of application of pressure to a predetermined temperature (the 2nd temperature) which is said under 1st temperature at least. Hereafter, this is called "cooling under application of pressure."

[0091] performing such cooling under application of pressure -- low [at the time of shaping] -- since a void content condition is maintained as it is, dimensional accuracy is high at a low void content, and the rare earth bond magnet which is excellent in magnetic properties is obtained.

[0092] It is desirable that it is the lowest possible temperature for reduction of the void content of the obtained bond magnet and improvement in dimensional accuracy, as for the 2nd temperature (decompressing temperature), it is desirable that they are the melting point of the thermoplastics used in this invention or the temperature not more than it, and it is more desirable that they are the heat deflection temperature (softening temperature) of the thermoplastics to be used or the temperature not more than it.

[0093] Moreover, as for the difference of said 1st temperature and 2nd temperature, it is desirable that it is 20 degrees C or more, and it is more desirable that it is 50 degrees C or more. Reduction of a void content and the effect of improvement in dimensional accuracy are so large that this temperature gradient is large.

[0094] In addition, when there are comparatively many contents of magnet powder, even if it sets up the 2nd temperature more highly, it is easy to obtain the bond magnet of a low void content. A void content can be made low (4.5% or less or 4.0% or less) also as the temperature near the melting point of the thermoplastics which follows, for example, uses the 2nd temperature when the content of the magnet powder in a kneading object is more than 94wt%, or a temperature more than the melting point (- melting point of about +10 degrees C).

[0095] Moreover, although cooling under application of pressure is good in a line once it cancels or eases the application of pressure at the time of pressing, it is desirable to be carried out continuously, without canceling the application of pressure at the time of pressing because of the simplification of a process, improvement in dimensional accuracy, etc.

[0096] Moreover, although the pressure in the case of cooling under application of pressure may be fixed or you may change, the melting point (especially heat deflection temperature) of the thermoplastics used at least of being held uniformly is desirable. When the pressure in the case of cooling under application of pressure changes, a pattern which increase or decrease in number continuously [a pressure] or gradually may be included.

[0097] Moreover, as for the pressure in the case of cooling under application of pressure (when this pressure carries out aging, it is the mean pressure), it is desirable that it is less than [the compacting pressure at the time of pressing, an EQC, or it], and it is more desirable that the melting point of the thermoplastics used at least is equivalent to the compacting pressure at the time of pressing. When also cooling the between from the melting point of the thermoplastics to be used to heat deflection temperature under application of pressure, as for a pressure in the meantime, it is desirable to carry out to about 40 - 100% of the compacting pressure at the time of pressing, and it is more desirable to consider as about 50 - 80%.

[0098] In addition, it cannot be overemphasized that cooling may be continued under pressureless (under ordinary pressure) after cooling under application of pressure (after decompressing) in this invention. Moreover, after performing cooling under pressureless, cooling under application of pressure may be

performed again.

[0099] Although especially the cooling rate in the case of cooling under application of pressure (it is the average when a cooling rate carries out aging) is not limited, it is desirable that it is 0.5–100 degrees C/second, and it is more desirable that it is 1–80 degrees C/second. When a cooling rate is too quick, by rapid contraction accompanying cooling, a detailed crack occurs, and a possibility of causing lowering of a mechanical strength is in the interior of a Plastic solid, and internal stress may increase by cooling, the strain and deformation by stress relaxation may arise at the time of the ** material from metal mold, and dimensional accuracy may fall. On the other hand, if a cooling rate is too slow, the cycle time of shaping will increase and productivity will fall.

[0100] Moreover, when continuing cooling also after decompressing, especially the cooling rate is not limited but can be made into the same cooling rate as the above.

[0101] In addition, the cooling rate in the case of cooling after decompressing in the case of cooling under application of pressure may be fixed, or may change, respectively.

[0102] In addition, in this process, what kind of method may be used for the method of cooling for the combination of an air cooling without blower, forced-air cooling, water cooling, oil quenching, water cooling, and air cooling etc.

[0103] The rare earth bond magnet manufactured by the method of above this inventions has the following outstanding properties. That is, it can take still more preferably for 2.0% or less 3.5% or less more preferably below 4.5% (vol%) preferably [a void content is low and]. Thus, by one with a low (= density is high) void content, dimensional accuracy is high, a mechanical strength is high and it excels in corrosion resistance, and also when it mass-produces, there is little variation in a size, and it excels in dimensional stability.

[0104] Furthermore, it excels in magnetic properties, and especially, from the numerousness of the presentation of magnet powder, and the contents of magnet powder etc., even if it is an isotropic magnet, it has outstanding magnetic properties.

[0105] That is, in the case of the rare earth bond magnet fabricated all over the non-magnetic field, it is maximum magnetic energy (product BH) max. In the case of the rare earth bond magnet which is 8 or more MGOes more preferably and was preferably fabricated all over the magnetic field 6 or more MGOes, it is maximum magnetic energy (product BH) max. Twelve or more MGOes are 13 or more MGOes more preferably.

[0106] in addition, the configuration of the rare earth bond magnet obtained by this invention, especially a size, etc. are limited -- not having -- for example, a configuration -- being related -- cylindrical, a prismatic form, and cylindrical -- the thing of all configurations, such as circular (imbricate form), plate-like, and bow tabular, is possible, and the thing of all magnitude is possible also for the magnitude from a large-sized thing to a micro thing.

[0107]

[Example] Hereafter, the concrete example of this invention is explained.

[0108] (Example 1) Mix magnet powder, following joint resin (thermoplastics), and a following additive, knead this mixture, and carry out the granulation (particle size regulation) of this kneading object. The granular object was obtained, it was filled up with this granular object in the metal mold of a making machine, and compression molding (blue heat forming) was carried out all over the non-magnetic field, and it cooled, maintaining the application-of-pressure condition at the time of shaping by this **, and the rare earth bond magnet (sample No.1–9) with which magnet powder was combined by solidification of joint resin was manufactured. In addition, each content of each matter shows the amount in a kneading object.

[0109] – configuration Nd-Fe-B system magnet powder: -- Nd12.0Fe77.8Co 4.3B5.9 and 96.0wt% -- A-G of a publication in the thermoplastics:table 1, an each 2.8wt% anti-oxidant:hydrazine system anti-oxidant, and a 1.2wt% mixing:Henschel mixer -- using -- mixing.

[0110] Kneading: Knead with a biaxial extrusion kneading machine. Kneading temperature is table 2 reference.

[0111] A screw speed 100 – 300rpm. Mixing-time 5 – 15 minute granulation (particle size regulation): Grinding and a classification adjust a kneading object to a grain with a mean particle diameter of 0.8mm.

[0112] Shaping: The granular object was fed into metal mold and pressing was carried out in the place heated to a predetermined molding temperature (the 1st temperature).

[0113] Refer to the table 2 for molding temperature and compacting pressure.

[0114] Cooling: It cooled to decompressing temperature (the 2nd temperature), maintaining an application-of-pressure condition, it cooled to ordinary temperature further after decompressing, and the sample was taken out.

[0115] The cooling method was made into air cooling. Decompressing temperature is table 2 reference.

[0116] The cooling rate in cooling under application of pressure is 1 degree C/second.

[0117] Mold-goods configuration: The shape of a cylindrical shape (an outer-diameter phi30mmx bore phi28mmx height of 7mm)

Plate configuration (3mm in 20mm angle x thickness) (for mechanical-strength measurement)

When the magnetic engine performance (flux density Br, coercive force iHc, and maximum magnetic energy (product BH) max), density, a void content, a mechanical strength, and corrosion resistance were investigated about the obtained rare earth bond magnet, it was as being shown in the following table 3.

[0118] In addition, assessment of each parameter in a table 3 followed the following methods.

[0119] Magnetic engine performance: 40kOe Maximum impression magnetic field 25kOe after carrying out pulse magnetization It measures with a direct-current magnetic-measurement machine. Or after starting a magnet piece with a 5mm angle x thickness of 1mm from a shaping sample, it measures by VSM.

[0120] Density: Measure by the Archimedes method (underwater method).

[0121] Void content: From the measured value of a weighing capacity presentation and the density of a Plastic solid to calculation

[0122] Mechanical strength: Measure by the punching shear test. Circular punch (outer diameter of 3mm) performed the testing machine by shear rate 1.0 mm/min using the autograph by Shimadzu Corp.

[0123] The magnet of a plate configuration is used for a sample.

[0124] corrosion-resistant : -- the constant temperature of the temperature of 80 degrees C, and 90% of humidity -- time amount until it feeds a shaping magnet into a constant humidity chamber and rust is generated on a magnet front face -- measurement. Surface observation is taken out from a tub every 50 hours, and is observed with an optical microscope (x10 time). 500 hours after observed every 500 hours.

[0125]

[A table 1]

樹脂 No.	結 合 樹 脂	融 点 [℃]	熱変形温度 [℃]
A	ポリアミド樹脂 (PA12)	178	145
B	ポリアミド樹脂 (コポリマPA6-12)	145	46
C	ポリアミド樹脂 (PA6)	215	180
D	ポリプロピレン樹脂 (PP)	174	105
E	ポリエチレン樹脂 (PE)	128	86
F	共重合ポリエステル	280	180
G	ポリフェニレンサルファイド (PPS)	287	260

[0126]

[A table 2]

(実施例1)

サンプル No.	結合樹脂	混練温度 [℃]	成形温度 [℃]	除圧温度 [℃]	成形圧力 [kgf/mm ²]
1	A	150~250	220	100	10
2	B	100~250	200	40	15
3	A (75%) + B (25%)	150~250	230	120	10
4	A (50%) + B (50%)	140~250	210	40	7.5
5	C	190~290	250	150	20
6	D	120~250	210	90	25
7	E	100~200	150	70	10
8	F	200~350	320	140	30
9	G	260~360	300	240	25

[0127]

[A table 3]

(実施例1)

サンプル No.	Br [kG]	iHc [kOe]	(BH) max [MGOe]	密度 [g/cm ³]	空孔率 [%]	機械的強度 [kgf/mm ²]	耐食性 [時間]
1	7.21	9.26	10.1	6.01	0.59	7.90	>500
2	7.19	9.31	10.0	6.03	0.73	7.45	>500
3	7.23	9.23	10.1	6.03	0.37	7.78	>500
4	7.22	9.21	10.0	6.03	0.49	7.60	>500
5	7.27	9.27	10.0	6.08	0.92	8.10	300
6	7.24	9.35	10.3	5.88	0.58	6.95	300
7	7.23	9.30	10.2	5.95	0.38	5.80	350
8	7.03	9.12	9.8	6.29	0.64	9.65	450
9	7.01	9.10	9.8	6.27	0.59	9.73	450

[0128] Although each rare earth bond magnet (sample No.1-9) by this invention using thermoplastics as joint resin was low compacting pressure, the void content was as low as 1% or less, and the bond magnet of the high density as theoretical density was obtained mostly, consequently it was able to obtain the magnet with a dramatically high mechanical strength, so that clearly from a table 3.

[0129] Moreover, it had corrosion resistance sufficient also in the condition of not performing coating for a magnet front face. It is presumed that this reason is because joint resin has covered the magnet powder front face to homogeneity according to there being few holes.

[0130] When observed by taking the electron microscope photograph (SEM) of the cutting plane about each magnet of sample No.1-9, most holes were not observed but it was checked that the joint resinous principle is distributing the circumference of magnet powder to homogeneity.

[0131] Furthermore, flux density Br and coercive force iHc The maximum magnetic energy product (BH) max It is high and it turns out that they are outstanding magnetic properties.

[0132] (Example 1 of a comparison) The granular object was obtained, following magnet powder and joint resin (thermosetting resin) were mixed, this mixture was kneaded, compression molding (cold forming or blue heat forming) was carried out [the granulation (particle size regulation) of this kneading object is carried out, / it was filled up with this granular object in the metal mold of a making machine,] all over the non-magnetic field, joint resin was stiffened after that, and the rare earth bond magnet (sample No.10-15) was manufactured. In addition, each content of each matter shows the amount in a kneading object.

[0133] - Configuration Nd-Fe-B system magnet powder : the thing of a publication in Nd_{12.0}Fe_{77.8} Co

4.3B5.9 and the 96.0wt% thermosetting resin:table 4, 4.0wt% (a curing agent is included)

Mixing: When solid resin is used at a room temperature, mix by the V shaped rotary mixer.

[0134] When liquefied resin is used at a room temperature, it mixes with an agitator.

[0135] Kneading: Knead using a kneader. Refer to the table 5 for kneading temperature.

[0136] The kneader rotational frequency 50 – 250rpm. Mixing time 30 minutes.

[0137] Granulation (particle size regulation): Grinding and a classification adjust a kneading object to a grain with a mean particle diameter of 0.8mm or less.

[0138] Shaping: The granular object was fed into metal mold and pressing was carried out with a predetermined molding temperature.

[0139] Refer to the table 5 for molding temperature and compacting pressure.

[0140] Cooling: It cooled to decompressing temperature (except for sample No.10 and 11), it cooled to ordinary temperature further after decompressing, and the sample was taken out.

[0141] The cooling method was made into air cooling. Decompressing temperature is table 5 reference.

[0142] A cooling rate is 2 degrees C/second.

[0143] Heat treatment: Put temporary mold goods into a thermostat and harden thermosetting resin.

[0144] Refer to the table 4 for hardening conditions.

[0145] Mold-goods configuration: The shape of a cylindrical shape (an outer-diameter phi30mmx bore phi28mmx height of 7mm)

Plate configuration (3mm in 20mm angle x thickness) (for mechanical-strength measurement)

When the magnetic engine performance (the maximum magnetic energy product (BH) max), density, a void content, a mechanical strength, and corrosion resistance were investigated about the obtained rare earth bond magnet, it was as being shown in the following table 6. In addition, the assessment method of each item is the same as that of an example 1.

[0146]

[A table 4]

樹脂 No.	結 合 樹 脂	軟化温度 [℃]	硬化条件
H	ビスフェノールA型エポキシ樹脂	室温以下	150℃×1hr
I	フェノールノボラック型エポキシ樹脂	80	170℃×2hr
J	フェノール樹脂	70	180℃×4hr

[0147]

[A table 5]

(比較例1)

サンプル No.	結 合 樹 脂	混 練 温 度 [℃]	成形温度 [℃]	除圧温度 [℃]	成形圧力 [kgf/mm ²]
10	H	室温	室温	室温	20
11	H	室温	室温	室温	70
12	I	80~100	120	50	20
13	I	80~100	120	50	70
14	J	70~90	100	50	20
15	J	70~90	100	50	70

[0148]

[A table 6]

(比較例1)

サンプル No.	(BH) max [MGOe]	密度 [g/cm ³]	空孔率 [%]	機械的強度 [kgf/mm ²]	耐食性 [時間]
10	8.2	5.70	7.7	測定不能	50
11	9.1	5.85	5.27	3.86	200
12	8.0	5.67	8.18	測定不能	50
13	9.0	5.82	5.75	3.91	200
14	8.2	5.73	9.51	測定不能	50
15	9.2	5.90	6.83	4.01	150

[0149] With the magnet (sample No.10-15) of the example of a comparison using thermosetting resin as joint resin so that clearly from a table 6, it is compacting pressure 20 kgf(s)/mm² When it carries out, of course, it is compacting pressure 70 kgf(s)/mm² Even when it carries out, a void content is high and the density of a magnet Plastic solid is low. Consequently, a magnetic mechanical strength is low and the corrosion resistance of a mechanical strength is also low.

[0150] When observed by taking the electron microscope photograph (SEM) of the cutting plane about each magnet of sample No.10-15, many holes existed in the interior. Moreover, distribution of the hole was the relation of the pressure transfer at the time of pressing, and the core had many holes, and it was in the uneven condition as [say / near a front face / that it is few]. Moreover, it was observed that the resinous principle is segregating.

[0151] Moreover, it is compacting pressure 20 kgf(s)/mm² When it carried out, the variation in the mechanical strength within a sample was large, therefore since [except the location to which the load was applied with circular punch] the crack and the crack were produced by the way, measurement of an exact mechanical strength was not completed. On the other hand, it is compacting pressure 70 kgf(s)/mm² When it carried out, the weld flash according [the resinous principle in a kneading object] to leakage and this occurred.

[0152] (Example 2) The granular object was obtained, the granulation (particle size regulation) of this kneading object is carried out, magnet powder, following joint resin (thermoplastics), and a following additive were mixed, this mixture was kneaded, and compression molding (blue heat forming) was carried out [it was filled up with this granular object in the metal mold of a making machine, and] all over the magnetic field, and it cooled, maintaining the application-of-pressure condition at the time of shaping by this **, and the rare earth bond magnet (sample No.16-19) was manufactured. In addition, each content of each matter shows the amount in a kneading object.

[0153] - Configuration Sm-Co system magnet powder : Sm (Cobal.Fe_{0.32}Cu_{0.06}Zr_{0.016})_{7.8}, 95.0wt% thermoplastics :P PS resin, a 4.2wt% anti-oxidant:hydrazine system anti-oxidant, 0.8wt% mixing: Mix using a V shaped rotary mixer.

[0154] Kneading: Use various kneading machines. Refer to the table 7 for kneading conditions.

[0155] Granulation (particle size regulation): Grinding and a classification adjust a kneading object to a grain with a mean particle diameter of 0.8mm.

[0156] Shaping: The granular object was fed into metal mold, and pressing was carried out in the place heated to a predetermined molding temperature (the 1st temperature), impressing a horizontal magnetic field (15kOe).

[0157] Molding temperature is 320 degrees C and compacting pressure is 20 kgf(s)/mm². It carried out.

[0158] Cooling: It cooled to the decompressing temperature (the 2nd temperature) of 150 degrees C, maintaining an application-of-pressure condition, it cooled to ordinary temperature further after decompressing, and the sample was taken out.

[0159] The cooling method was made into air cooling.

[0160] The cooling rate in cooling under application of pressure is 5 degrees C/second.

[0161] Mold-goods configuration: Rectangular parallelepiped (a 8mm[11mm by] x height of 7mm and the height direction are the direction of orientation.)

Plate configuration (3mm in 20mm angle x thickness) (for mechanical-strength measurement)

When the magnetic engine performance (the maximum magnetic energy product (BH) max), density, a void content, a mechanical strength, and corrosion resistance were investigated about the obtained rare earth bond magnet, it was as being shown in the following table 8. In addition, the assessment method of each item is the same as that of an example 1.

[0162]

[A table 7]

(実施例2)

サンプル No.	混練機	混練温度 [℃]	回転数 [rpm]	処理量
16	二軸押出混練機	170~320	100~250	30kg/hr
17	ロール混練機	180~300	10~100	5kg/batch, 15min/batch
18	ニーダー	180~300	20~100	10kg/batch, 30min/batch
19	KCK	170~320	20~80	20kg/hr

[0163]

[A table 8]

(実施例2)

サンプル No.	(BH) max [MGoe]	密度 [g/cm ³]	空孔率 [%]	機械的強度 [kgf/mm ²]	耐食性 [時間]
16	15.2	6.63	0.65	8.14	>1000
17	15.5	6.65	0.35	8.23	>1000
18	14.9	6.61	0.95	8.09	>1000
19	15.3	6.63	0.65	8.19	>1000

[0164] The void content was as low as 1% or less, the bond magnet of high density was obtained, consequently each rare earth bond magnet (sample No.16-19) by this invention had a mechanical strength and high corrosion resistance so that clearly from a table 8.

[0165] Moreover, when observed by taking an electron microscope photograph (SEM) like the above about each magnet of sample No.16-19, most holes were not observed but it was checked that the joint resinous principle is distributing the circumference of magnet powder to homogeneity.

[0166] Furthermore, maximum magnetic energy (product BH) max It is high and it turns out that they are outstanding magnetic properties.

[0167] (Example 2 of a comparison) Magnet powder, following joint resin (thermoplastics), and a following additive were mixed, it was filled up with this mixture in the metal mold of a making machine, compression molding (blue heat forming) was carried out all over the magnetic field, and the rare earth bond magnet (sample No.20, 21) was manufactured. In addition, each content of each matter shows the amount in mixture.

[0168] - Configuration Sm-Co system magnet powder : Sm (Cobal.Fe0.32Cu0.06Zr0.016)7.8, 95.0wt% (sample No.20), and 96.0wt% (sample No.21)

Thermoplastics :P PS resin, 4.2wt% (sample No.20), and 3.2wt% (sample No.21)

Anti-oxidant: Mix using a hydrazine system anti-oxidant and a 0.8wt% mixing:V shaped rotary mixer.

[0169] Shaping: Mixture was fed into metal mold, and pressing was carried out in the place heated to a

predetermined molding temperature, impressing a horizontal magnetic field (15kOe).

[0170] Molding temperature is 320 degrees C and compacting pressure is 2 20 kgf(s)/mm. It carried out.

[0171] Cooling: It cooled to the temperature of 150 degrees C, and the sample was taken out.

[0172] The cooling method was made into air cooling.

[0173] A cooling rate is 5 degrees C/second.

[0174] Mold-goods configuration: Rectangular parallelepiped (a 8mm[11mm by] x height of 7mm and the height direction are the direction of orientation.)

Plate configuration (3mm in 20mm angle x thickness) (for mechanical-strength measurement)

By reaching sample No.20, when resin leakage arose at the time of shaping and the edge and the end-face section of mold goods adhered to punch of a making machine, ***** of mold goods arose, and each of magnets of 21 could acquire those, such as an edge, or the configuration of injury student ***** and a request, and cut it.

[0175] When observed by taking the fabricated electron microscope photograph (SEM) of a portion, distribution of a joint resinous principle had become an ununiformity and was in the condition in which the magnet powder portion and the joint resin portion were intermingled.

[0176] Moreover, as mentioned above, it reached sample No.20, and since each of magnets of 21 was defectives, effective measurement of a mechanical strength etc. was not completed.

[0177] (Example 3) Mix magnet powder (two sorts), following joint resin (thermoplastics), and a following additive, knead this mixture, and carry out the granulation (particle size regulation) of this kneading object. The granular object was obtained, it was filled up with this granular object in the metal mold of a making machine, and compression molding (blue heat forming) was carried out all over the magnetic field, and it cooled, maintaining the application-of-pressure condition at the time of shaping by this **, and the rare earth bond magnet (sample No.22-30) was manufactured. In addition, each content of each matter shows the amount in a kneading object.

[0178] - Configuration Sm-Co system magnet powder : mix using Sm (Co0.672 Fe0.22Cu0.08Zr0.028)8.35, 70.5wt%Sm-Fe-N system magnet powder:Sm2 Fe17N3, 23.5wt% thermoplastics:polyamide resin (Nylon 12), a 5.0wt% antioxidant:phenolic antioxidant, and a 1.0wt% mixing:Henschel mixer.

[0179] Kneading: Knead with a biaxial extrusion kneading machine. Kneading temperature is 150-300 degrees C.

[0180] A screw speed 100 - 300rpm. Mixing-time 10 minute granulation (particle size regulation): Adjust a kneading object to the grain size shown in a table 9 by grinding and the classification.

[0181] Shaping: The granular object was cut by rubbing and it supplied to metal mold by the method, and pressing was carried out in the place heated at 220 degrees C (the 1st temperature), impressing a horizontal magnetic field (15kOe). Compacting pressure is 2 10 kgf(s)/mm. It carried out.

[0182] Cooling: It cooled to the decompressing temperature (the 2nd temperature) of 100 degrees C, maintaining an application-of-pressure condition, and the sample was taken out.

[0183] The cooling method was made into water cooling.

[0184] The cooling rate in cooling under application of pressure is 20 degrees C/second.

[0185] Mold-goods configuration: Plate configuration (a 2.5mm[in width-of-face / of 15mm / x thickness] x height of 5mm and the height direction are the direction of orientation)

When magnetic weight, density, a void content, and height were measured about the obtained rare earth bond magnet, it was as being shown in the following table 9.

[0186]

[A table 9]

(実施例3)

サンプル No.	粒状物の平均 粒径 [mm]	成形品重量 [mg]	密度 [g/cm ³]	空孔率 [%]	高さ [mm]
22	2	1073	5.78	0.30	4.95
23	1.8	1075	5.78	0.30	4.96
24	1.5	1077	5.78	0.30	4.97
25	1	1079	5.78	0.30	4.98
26	0.5	1083	5.79	0.12	4.99
27	0.1	1081	5.79	0.12	4.98
28	0.05	1080	5.76	0.64	5.00
29	0.01	1075	5.72	1.33	5.01
30	0.007	1071	5.68	2.02	5.03

注) 成形品重量と高さは、n=10の平均値

[0187] By setting out of the particle size of a granular object, the outstanding quantum nature is obtained, it is a low void content and a bond magnet with high dimensional accuracy is obtained so that clearly from a table 9. Especially, when the particle size of a granular object was the range which is 0.01–2mm, it could be compatible with a super-low void content (1% or less) and close dimensional accuracy (a size error is less than $\pm 5/100$ mm).

[0188] (An example 4, example 3 of a comparison) Mix magnet powder, following joint resin (thermoplastics), and a following additive, knead this mixture, and carry out the granulation (particle size regulation) of this kneading object. The granular object was obtained, it was filled up with this granular object in the metal mold of a making machine, and compression molding (blue heat forming) was carried out all over the magnetic field, and it cooled, maintaining the application-of-pressure condition at the time of shaping by this **, and the rare earth bond magnet (sample No.31–42) was manufactured. In addition, each content of each matter shows the amount in a kneading object.

[0189] – configuration Nd–Fe–B system magnet powder: — Nd_{12.6}Fe_{69.3}Co_{12.0}B_{6.0}Zr_{0.1} and 97.0wt% thermoplastics: — A or F in a table 1, an each 1.5wt% anti-oxidant:hydrazine system anti-oxidant, 1.4wt% lubricant:zinc stearate, and a 0.1wt% mixing:Henschel mixer — using — mixing.

[0190] Kneading: Knead with a biaxial extrusion kneading machine. Kneading temperature is 150–350 degrees C.

[0191] A screw speed 100 – 300rpm. Mixing-time 5 minute granulation (particle size regulation): Grinding and a classification adjust a kneading object to a grain with a mean particle diameter of 0.3mm.

[0192] Shaping: The granular object was fed into metal mold, and pressing was carried out in the place heated to the molding temperature (the 1st temperature) shown in a table 10, impressing a radial magnetic field (15kOe). Compacting pressure is 2 15 kgf(s)/mm. It carried out.

[0193] Cooling: It cooled to the decompressing temperature (the 2nd temperature) of 100 degrees C, maintaining an application-of-pressure condition, it cooled to ordinary temperature further after decompressing, and the sample was taken out.

[0194] The cooling method was made into water cooling.

[0195] The cooling rate in cooling under application of pressure is 30 degrees C/second.

[0196] Mold-goods configuration: The shape of a cylindrical shape (it pressurizes in an outer-diameter phi20mmx bore phi18mmx height of 5mm, and the height direction)

Plate configuration (3mm in 20mm angle x thickness) (for mechanical-strength measurement)

When the magnetic engine performance (the maximum magnetic energy product (BH) max), density, the void content, and the mechanical strength were investigated about the obtained rare earth bond magnet (example 4: sample No.32–36, 38–42, example of comparison 3:sample No.31, 37), it was as being shown in the following table 10. In addition, the assessment method of each item is the same as that of an example 1.

[0197]

[A table 10]

(実施例4、比較例3)

サンプル No.	結合樹脂	成形温度 [℃]	(BH) max [MG0e]	密 度 [g/cm ³]	空 孔 率 [%]	機械的強度 [kgf/mm ²]
31	A	130	測定不能	測定不能	測定不能	測定不能
32	A	150	17.0	6.21	2.52	5.10
33	A	180	17.5	6.28	1.42	7.10
34	A	200	18.2	6.34	0.48	7.70
35	A	300	17.8	6.32	0.79	7.61
36	A	360	16.2	6.32	0.79	7.55
37	F	160	測定不能	測定不能	測定不能	測定不能
38	F	190	16.9	6.36	2.70	8.10
39	F	250	17.5	6.44	1.48	9.25
40	F	300	18.0	6.50	0.56	9.78
41	F	350	17.6	6.50	0.56	9.65
42	F	400	15.5	6.50	0.56	9.60

[0198] Like sample No.32-36 in a table 10, and 38-42 (example 4), when molding temperature was more than the heat deflection temperature of joint resin, joint resin was able to change into softening or a melting condition at the time of shaping, and it was able to fabricate.

[0199] Especially, like sample No.33-36, and 40 and 42, when molding temperature is more than the melting point of joint resin, the void content of the obtained magnet decreases further and becomes higher [the magnetic engine performance].

[0200] On the other hand, since joint resin did not become soft like sample No.31 and 37 (example 3 of a comparison) at the time of shaping when molding temperature is under the heat deflection temperature of joint resin, a granular object could not fix mutually, a configuration could not be held, but shaping impossible or shaping was faulty. Therefore, measurement was impossible also about each parameter.

[0201] (An example 5, example 4 of a comparison) Mix magnet powder, following joint resin (thermoplastics), and a following additive, knead this mixture, and carry out the granulation (particle size regulation) of this kneading object. The granular object was obtained, it was filled up with this granular object in the metal mold of a making machine, and compression molding (blue heat forming) was carried out all over the non-magnetic field, and it cooled, maintaining the application-of-pressure condition at the time of shaping by this **, and the rare earth bond magnet (sample No.43-52) was manufactured. In addition, each content of each matter shows the amount in a kneading object.

[0202] - configuration nano crystal Nd-Fe-B system magnet powder: -- A or G in Nd5.5 Fe66B18.5Co5 Cr5 and the 98.0wt% thermoplastics:table 1, and each 1.0wt% anti-oxidant:hydrazine system anti-oxidant 1.0wt% mixing: -- a Henschel mixer -- using -- mixing.

[0203] Kneading: Knead with a biaxial extrusion kneading machine. Kneading temperature is 150-350 degrees C.

[0204] A screw speed 100 - 300rpm. Mixing-time 10 minute granulation (particle size regulation): Grinding and a classification adjust a kneading object to a grain with a mean particle diameter of 0.1mm.

[0205] Shaping: The granular object was fed into metal mold and pressing was carried out in the place heated to a predetermined molding temperature (the 1st temperature). For molding temperature, 200 degrees C (resin A) and 300 degrees C (resin G), and compacting pressure are 2 25 kgf(s)/mm. It carried out.

[0206] Cooling: Maintaining an application-of-pressure condition, it cooled to the decompressing temperature (the 2nd temperature) shown in a table 11, and the sample was taken out. The cooling method was made into water cooling.

[0207] The cooling rate in cooling under application of pressure is 50 degrees C/second.

[0208] Mold-goods configuration: The shape of a cylindrical shape (it pressurizes in an outer-diameter phi10mmx bore phi7mmx height of 7mm, and the height direction)

When the magnetic engine performance (the maximum magnetic energy product (BH) max), density, the void content, and the outer diameter were investigated about the obtained rare earth bond magnet (example 5: sample No.44-47, 49-52, example of comparison 4:sample No.43, 48), it was as being shown in the following table 11. In addition, the assessment method of each item is the same as that of an example 1.

[0209]

[A table 11]

(実施例5、比較例4)

サンプル No.	結合樹脂	除圧温度 [℃]	(BH) max [MG0e]	密度 [g/cm ³]	空孔率 [%]	外 径 [mm]
43	A	200	8.0	6.49	3.55	10.00±0.08
44	A	180	8.2	6.56	2.51	10.00±0.05
45	A	160	8.6	6.65	1.17	10.00±0.02
46	A	140	8.6	6.67	0.87	10.01±0.02
47	A	100	8.7	6.68	0.73	10.01±0.01
48	G	300	7.3	6.55	4.26	10.04±0.10
49	G	275	7.9	6.64	2.95	10.02±0.04
50	G	260	8.3	6.74	1.49	10.01±0.03
51	G	240	8.5	6.78	0.90	10.00±0.01
52	G	200	8.5	6.79	0.76	10.00±0.01

注) 外径は、n=10の測定値から算出

[0210] The void content of the magnet with which decompressing temperature was acquired when the difference of below the melting point of joint resin or decompressing temperature, and molding temperature was 20 degrees C or more is low like sample No.44-47 in a table 11, and 49-52 (example 5), density is high, the magnetic engine performance is high, and dimensional accuracy is high (a size error is less than **5/100mm). Such a property is improving, so that decompressing temperature is low.

[0211] Especially, like sample No.46, and 47, 50 and 51, when decompressing temperature is below the heat deflection temperature of joint resin, the density almost near theoretical density can be attained and it becomes the magnet of the extremely excellent magnetic engine performance which can fully demonstrate the property of magnet powder.

[0212] On the other hand, like sample No.43 and 48 (example 4 of a comparison), when decompressing temperature and molding temperature are the same, dimensional accuracy is low and a void content is also high compared with said sample No.44-47, and 49-52.

[0213]

[Effect of the Invention] The rare earth bond magnet which it excelled in the moldability also in the amount of little joint resin performing pressing by blue heat forming using the granular object of a kneading object according to [as stated above] this invention, and by cooling in the state of application of pressure after blue heat forming and to predetermined temperature further, and the mechanical strength was high at the low void content, and whose dimensional stability (dimensional accuracy) was high, and was excellent in magnetic properties can be offered.

[0214] In this case, when the particle size of a granule is the range of desired, a void content is very low and, moreover, dimensional stability improves further.

[0215] Moreover, since it carries out pressing, using thermoplastics as softening or a melting condition, the rare earth bond magnet of the above-mentioned property can be manufactured with comparatively low compacting pressure, and manufacture is easy.

[0216] The temperature below the melting point of the thermoplastics which the 2nd temperature at the time of cooling (decompressing temperature) uses especially, when it is the temperature below heat

deflection temperature further, or when having deviated the 1st temperature and beyond predetermined temperature, a rare earth bond magnet with very high dimensional stability with a very low and void content can be offered.

[Translation done.]



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(54) 【発明の名称】 希土類ボンド磁石の製造方法

(57) 【要約】

【課題】 空孔率が低く、寸法精度が高く、成形性、磁気特性に優れた希土類ボンド磁石を提供すること。

【解決手段】 本発明の希土類ボンド磁石の製造方法は、まず、希土類磁石粉末と、熱可塑性樹脂よりなる結合樹脂と、酸化防止剤とを所定の比率で混合し、これを前記熱可塑性樹脂の熱変形温度以上の温度で混練し、次いで、該混練物を造粒または整粒し、得られた粒状物を用いて、前記熱可塑性樹脂が軟化または熔融状態となる第1の温度で加圧成形を行い、その後、少なくとも前記第1の温度未満である第2の温度まで加圧状態で冷却し、熱可塑性樹脂を固化することにより希土類ボンド磁石を製造するものである。第2の温度は、用いる熱可塑性樹脂の融点以下または熱変形温度以下であるのが好ましい。

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【特許請求の範囲】

【請求項1】 希土類磁石粉末を熱可塑性樹脂よりなる結合樹脂により結合してなる希土類ボンド磁石の製造方法であって、

前記希土類磁石粉末と前記結合樹脂とを混合・混練して混練物を製造する工程と、

前記混練物を造粒または整粒して粒状物とする工程と、
前記粒状物を用いて前記結合樹脂が軟化または熔融状態となる第1の温度で加圧成形する工程と、

少なくとも前記第1の温度未満である第2の温度まで加圧状態で冷却する工程とを有することを特徴とする希土類ボンド磁石の製造方法。

【請求項2】 前記混練は、前記結合樹脂の熱変形温度以上の温度で、かつ前記希土類磁石粉末の表面が熔融または軟化した結合樹脂成分により覆われた状態となるように行われる請求項1に記載の希土類ボンド磁石の製造方法。

【請求項3】 前記混練物中の前記希土類磁石粉末の含有量が90～99wt%である請求項1または2に記載の希土類ボンド磁石の製造方法。

【請求項4】 前記混練物中に、酸化防止剤を含有する請求項1ないし3のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項5】 前記混練物中の前記酸化防止剤の含有量が0.1～2wt%である請求項4に記載の希土類ボンド磁石の製造方法。

【請求項6】 前記造粒または整粒は、粉碎により行われる請求項1ないし5のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項7】 前記粒状物の平均粒径が10 μ m～2mmである請求項1ないし6のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項8】 前記加圧成形は、圧縮成形である請求項1ないし7のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項9】 前記第2の温度は、前記結合樹脂の融点である請求項1ないし8のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項10】 前記第2の温度は、前記結合樹脂の熱変形温度である請求項1ないし8のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項11】 前記第1の温度と前記第2の温度との差が、20℃以上である請求項1ないし10のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項12】 前記加圧状態での冷却は、前記加圧成形の際の加圧を解除することなく連続して行われる請求項1ないし11のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項13】 前記加圧成形時の成形圧力に対し、前記加圧状態での冷却時の圧力が同等またはそれ以下であ

る請求項1ないし12のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項14】 前記加圧状態での冷却時の圧力は、少なくとも前記結合樹脂の融点まで一定に保持されている請求項1ないし13のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項15】 前記加圧状態での冷却時の圧力は、少なくとも前記第1の温度と第2の温度の間の温度まで一定に保持されている請求項1ないし13のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項16】 前記加圧状態での冷却時の冷却速度は、0.5～100℃/秒である請求項1ないし15のいずれかに記載の希土類ボンド磁石の製造方法。

【請求項17】 前記加圧成形時の成形圧力は、60kgf/mm²以下である請求項1ないし16のいずれかに記載の希土類ボンド磁石の製造方法。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、希土類ボンド磁石の製造方法および希土類ボンド磁石用組成物に関するものである。

【0002】

【従来の技術】希土類ボンド磁石は、希土類磁石粉末と結合樹脂（有機バインダー）との混合物（コンパウンド）を所望の磁石形状に加圧成形して製造されるものである。この加圧成形による成形方法には、大別して、圧縮成形法、射出成形法および押出成形法がある。

【0003】圧縮成形法は、前記コンパウンドをプレス金型中に充填し、これを所定温度で圧縮成形して成形体を得、その後、結合樹脂が熱硬化性樹脂である場合にはそれを硬化させて磁石とする方法である。この方法は、他の方法に比べ、結合樹脂の量が少なくても成形が可能であるため、得られた磁石中の樹脂量が少なくなり、磁気特性の向上にとって有利である。

【0004】押出成形法は、加熱熔融された前記コンパウンドを押出成形機の金型から押し出すとともに冷却固化し、所望の長さに切断して、磁石とする方法である。この方法では、磁石の形状に対する自由度が大きく、薄肉、長尺の磁石をも容易に製造できるという利点があるが、成形時における熔融物の流動性を確保するために、結合樹脂の添加量を圧縮成形法のそれに比べて多くする必要があり、従って、得られた磁石中の樹脂量が多く、磁気特性が低下する傾向があるという欠点がある。

【0005】射出成形法は、前記コンパウンドを加熱熔融し、十分な流動性を持たせた状態で該熔融物を金型内に注入し、所定の磁石形状に成形する方法である。この方法では、磁石の形状に対する自由度は、押出成形法に比べさらに大きく、特に、異形状の磁石をも容易に製造できるという利点がある。しかし、成形時における熔融物の流動性は、前記押出成形法より高いレベルが要求さ

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れるので、結合樹脂の添加量は、押出成形法のそれに比べてさらに多くする必要があり、従って、得られた磁石中の樹脂量が多く、磁気特性が低下する傾向があるという欠点がある。

【0006】

【発明が解決しようとする課題】ところで、以上のような各方法の内、最も磁気性能の高い磁石を成形可能な圧縮成形には、次のような欠点がある。

【0007】第1に、製造された希土類ボンド磁石は、成形体の密度は高いものの空孔率が高くなる傾向を示すため、機械的強度が弱く、耐食性に劣る。そのため、特に圧縮成形法においては、成形圧力を 70 kgf/mm^2 以上と高圧にする高圧成形を活用したり、成形後に防食用コーティング処理を施す等の方法で対処していた。しかしながら、高圧成形は、成形機への負担が大きく、また、防食用コーティング処理を行う場合には、そのための工程が追加され、製造工程の複雑化による生産性の低下、製造コストの上昇を招く。

【0008】第2に、熱硬化性樹脂を使用したコンパウンドは、樹脂が未硬化状態であるため、樹脂の硬化による物性の変化や吸水による物性の変化が生じ、コンパウンドの成形性が経時的に変化する。これにより、同一条件で成形した場合でも成形体の寸法や密度が変わり、安定に成形を行うことが困難となる。また、熱硬化性樹脂を使用した場合には、キュアリング（硬化）工程が必要になり、これによって工程の増加やコストアップにつながるだけでなく、キュアリング時の樹脂の反応による寸法の変化が生じ、目標寸法を確保するためには、金型寸法の補正が必要になり、寸法確保が容易でない。

【0009】また、従来の圧縮成形の場合、熱硬化性樹脂として室温で固体のものと液状のもの両者が使用される。このうち、前者の固体樹脂を用いたときには、給材性は比較的良好ではあるが成形性が悪く、より空孔率が高くなる傾向を示す。また、樹脂と磁石粉末の分散性が悪く、その結果、機械的強度が低下する。一方、後者の液状樹脂を用いたときには、高密度の成形体を得ることは可能であるが、成形時の環境（温度、湿度）による影響で敏感に樹脂の物性が変化し、金型への充填性が低下する。

【0010】そのため、磁石の目標寸法に対しバラツキが生じ、すなわち寸法精度が悪く、成形の安定性に欠ける。特に、小型の磁石の場合には、この欠点は顕著となる。このように寸法のバラツキが大きいことから、最終磁石製品の目標寸法を確保するために、目標寸法よりも大きく成形した後、切削・研磨等の二次加工により寸法を調整する必要がある。これにより、工程の増加を招き、加工により不良材料が発生するので、生産性が低下し、製造コストが増加する。また、このような欠点を解消するためには、成形機の構造や成形工程に特殊な工夫を施さねばならず、成形機の消耗も著しく、成形のサイ

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クルタイムも長くなる。

【0011】さらに、以上のような第1および第2の欠点は、コンパウンドの製造方法、製造条件、成形時の温度条件、成形後の冷却条件等が不適切であるのも原因の一つとなっている。

【0012】従って、本発明の目的は、成形性、磁気特性、寸法安定性に優れた低空孔率の希土類ボンド磁石を容易に製造することができる希土類ボンド磁石の製造方法を提供することにある。

10 【0013】

【課題を解決するための手段】このような目的は、下記（1）～（17）の本発明により達成される。

【0014】（1）希土類磁石粉末を熱可塑性樹脂よりなる結合樹脂により結合してなる希土類ボンド磁石の製造方法であって、前記希土類磁石粉末と前記結合樹脂とを混合・混練して混練物を製造する工程と、前記混練物を造粒または整粒して粒状物とする工程と、前記粒状物を用いて前記結合樹脂が軟化または溶融状態となる第1の温度で加圧成形する工程と、少なくとも前記第1の温度未満である第2の温度まで加圧状態で冷却する工程とを有することを特徴とする希土類ボンド磁石の製造方法。

【0015】（2）前記混練は、前記結合樹脂の熱変形温度以上の温度で、かつ前記希土類磁石粉末の表面が溶融または軟化した結合樹脂成分により覆われた状態となるように行われる上記（1）に記載の希土類ボンド磁石の製造方法。

【0016】（3）前記混練物中の前記希土類磁石粉末の含有量が $90\sim 99\text{ wt}\%$ である上記1または2に記載の希土類ボンド磁石の製造方法。

【0017】（4）前記混練物中に、酸化防止剤を含有する上記（1）ないし（3）のいずれかに記載の希土類ボンド磁石の製造方法。

【0018】（5）前記混練物中の前記酸化防止剤の含有量が $0.1\sim 2\text{ wt}\%$ である上記（4）に記載の希土類ボンド磁石の製造方法。

【0019】（6）前記造粒または整粒は、粉碎により行われる上記（1）ないし（5）のいずれかに記載の希土類ボンド磁石の製造方法。

【0020】（7）前記粒状物の平均粒径が $10\mu\text{m}\sim 2\text{ mm}$ である上記（1）ないし（6）のいずれかに記載の希土類ボンド磁石の製造方法。

【0021】（8）前記加圧成形は、圧縮成形である上記（1）ないし（7）のいずれかに記載の希土類ボンド磁石の製造方法。

【0022】（9）前記第2の温度は、前記結合樹脂の融点である上記（1）ないし（8）のいずれかに記載の希土類ボンド磁石の製造方法。

【0023】（10）前記第2の温度は、前記結合樹脂の熱変形温度である上記（1）ないし（8）のいずれか

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に記載の希土類ボンド磁石の製造方法。

【0024】(11) 前記第1の温度と前記第2の温度との差が、20℃以上である上記(1)ないし(10)のいずれかに記載の希土類ボンド磁石の製造方法。

【0025】(12) 前記加圧状態での冷却は、前記加圧成形の際の加圧を解除することなく連続して行われる上記(1)ないし(11)のいずれかに記載の希土類ボンド磁石の製造方法。

【0026】(13) 前記加圧成形時の成形圧力に対し、前記加圧状態での冷却時の圧力が同等またはそれ以下である上記(1)ないし(12)のいずれかに記載の希土類ボンド磁石の製造方法。

【0027】(14) 前記加圧状態での冷却時の圧力は、少なくとも前記結合樹脂の融点まで一定に保持されている上記(1)ないし(13)のいずれかに記載の希土類ボンド磁石の製造方法。

【0028】(15) 前記加圧状態での冷却時の圧力は、少なくとも前記第1の温度と第2の温度の間の温度まで一定に保持されている上記(1)ないし(13)のいずれかに記載の希土類ボンド磁石の製造方法。

【0029】(16) 前記加圧状態での冷却時の冷却速度は、0.5～100℃/秒である上記(1)ないし(15)のいずれかに記載の希土類ボンド磁石の製造方法。

【0030】(17) 前記加圧成形時の成形圧力は、60kgf/mm²以下である上記(1)ないし(16)のいずれかに記載の希土類ボンド磁石の製造方法。

【0031】

【発明の実施の形態】以下、本発明の希土類ボンド磁石の製造方法について詳細に説明する。

【0032】本発明の希土類ボンド磁石の製造方法は、主に、以下の工程を有している。

【0033】<1>希土類ボンド磁石用組成物の混練物の製造

まず、希土類ボンド磁石用組成物（以下単に「組成物」と言う）を調整する。この組成物は、主に、希土類磁石粉末と、結合樹脂（バインダー）とで構成される。また、好ましくは酸化防止剤を含有し、必要に応じその他の添加剤が添加される。これらの各構成成分は、例えば、ヘンシェルミキサー等の混合機や攪拌機を用いて混合され、さらに、後述するように混練されて混練物を得る。

【0034】以下、これらの各構成成分について説明する。

【0035】1. 希土類磁石粉末

希土類磁石粉末としては、希土類元素と遷移金属とを含む合金よりなるものが好ましく、特に、次の【1】～

【4】が好ましい。

【0036】【1】 Smを主とする希土類元素と、Coを主とする遷移金属とを基本成分とするもの（以下、

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Sm-Co系合金と言う）。

【0037】【2】 R（ただし、RはYを含む希土類元素のうち少なくとも1種）と、Feを主とする遷移金属と、Bとを基本成分とするもの（以下、R-Fe-B系合金と言う）。

【0038】【3】 Smを主とする希土類元素と、Feを主とする遷移金属と、Nを主とする格子間元素とを基本成分とするもの（以下、Sm-Fe-N系合金と言う）。

【0039】【4】 R（ただし、RはYを含む希土類元素のうち少なくとも1種）とFe等の遷移金属とを基本成分とし、ナノメートルレベルで磁性相を有するもの（以下、「ナノ結晶磁石」と言う）。

【0040】【5】 前記【1】～【4】の組成のものうち、少なくとも2種を混合したもの。この場合、混合する各磁石粉末の利点を併有することができ、より優れた磁気特性を容易に得ることができる。

【0041】Sm-Co系合金の代表的なものとしては、SmCo₅、Sm₂TM₁₇（ただしTMは、遷移金属）が挙げられる。

【0042】R-Fe-B系合金の代表的なものとしては、Nd-Fe-B系合金、Pr-Fe-B系合金、Nd-Pr-Fe-B系合金、Ce-Nd-Fe-B系合金、Ce-Pr-Nd-Fe-B系合金、これらにおけるFeの一部をCo、Ni等の他の遷移金属で置換したもの等が挙げられる。

【0043】Sm-Fe-N系合金の代表的なものとしては、Sm₂Fe₁₇合金を窒化して作製したSm₂Fe₁₇N₃が挙げられる。

【0044】磁石粉末における前記希土類元素としては、Y、La、Ce、Pr、Nd、Pm、Sm、Eu、Gd、Tb、Dy、Ho、Er、Tm、Yb、Lu、ミッシュメタルが挙げられ、これらを1種または2種以上含むことができる。また、前記遷移金属としては、Fe、Co、Ni等が挙げられ、これらを1種または2種以上含むことができる。また、磁気特性を向上させるために、磁石粉末中には、必要に応じ、B、Al、Mo、Cu、Ga、Si、Ti、Ta、Zr、Hf、Ag、Zn等を含有することもできる。

【0045】また、磁石粉末の平均粒径は、特に限定されないが、0.5～100μm程度が好ましく、1～50μm程度がより好ましい。なお、磁石粉末等の粒径は、例えば、F.S.S.S.（Fischer Sub-Sieve Sizer）法により測定することができる。

【0046】また、磁石粉末の粒径分布は、均一でも、ある程度分散されていても（バラツキがある）よいが、後述するような少量の結合樹脂で成形時の良好な成形性を得るために、後者の方が好ましい。これにより、得られたボンド磁石の空孔率をより低減することもできる。

なお、前記【5】の場合、混合する磁石粉末の組成毎

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に、その平均粒径が異なってもよい。

【0047】磁石粉末の製造方法は、特に限定されず、例えば、溶解・ casting により合金インゴットを作製し、この合金インゴットを適度な粒度に粉碎し（さらに分級し）て得られたもの、アモルファス合金を製造するのに用いる急冷薄帯製造装置で、リボン状の急冷薄片（微細な多結晶が集合）を製造し、この薄片（薄帯）を適度な粒度に粉碎し（さらに分級し）て得られたもの等、いずれでもよい。

【0048】以上のような磁石粉末の混練物中での含有量は、90～99wt%程度であるのが好ましく、93～99wt%程度であるのがより好ましく、95～99wt%程度であるのがより好ましい。磁石粉末の含有量が少な過ぎると、磁気特性（特に磁気エネルギー積）の向上が図れず、また、磁石粉末の含有量が多過ぎると、相対的に結合樹脂の含有量が少なくなり、成形性が低下する。

【0049】2. 結合樹脂（バインダー）

結合樹脂（バインダー）としては、熱可塑性樹脂が用いられる。結合樹脂として熱可塑性樹脂を用いた場合には、熱硬化性樹脂を用いた場合に比べ、低空孔率の磁石を得る上で有利であるが、本発明では、後述する成形時の温度条件、冷却条件と相まって、より低い空孔率を実現することができる。

【0050】熱可塑性樹脂としては、例えば、ポリアミド（例：ナイロン6、ナイロン46、ナイロン66、ナイロン610、ナイロン612、ナイロン11、ナイロン12、ナイロン6-12、ナイロン6-66）、熱可塑性ポリイミド、芳香族ポリエステル系樹脂等の液晶ポリマー、ポリフェニレンオキシド、ポリフェニレンサルファイド、ポリエチレン、ポリプロピレン、エチレン-酢酸ビニル共重合体等のポリオレフィン、変性ポリオレフィン、ポリエーテル、ポリアセタール等、またはこれらを主とする共重合体、ブレンド体、ポリマーアロイ等が挙げられ、これらのうちの1種または2種以上を混合して用いることができる。

【0051】これらのうちでも、成形性に優れ、また機械的強度が強いことから、ポリアミドまたはその共重合体、耐熱性向上の点から、液晶ポリマー、ポリフェニレンサルファイドを主とするもの、成形の容易性や低コストの点で、ポリオレフィンを主とするものが好ましい。また、これらの熱可塑性樹脂は、磁石粉末との混練性にも優れている。

【0052】用いられる熱可塑性樹脂は、融点が120℃以上のものであるのが好ましく、122℃～400℃のものであるのがより好ましく、125℃～350℃のものであるのがさらに好ましい。融点が前記下限値未満のものであると、磁石成形体の耐熱性が低下し、十分な温度特性（磁氣的または機械的）を確保することが困難となる。また、融点が前記上限値を超えるものであると、成形時の温度が上昇し、磁石粉末等の酸化が生じ易

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くなる。

【0053】また、成形性をより向上させるために、用いられる熱可塑性樹脂の平均分子量（重合度）は、10000～60000程度であるのが好ましく、10000～35000程度であるのがより好ましい。

【0054】以上のような結合樹脂の混練物中での含有量は、1～10wt%程度であるのが好ましく、1～8wt%程度であるのが好ましく、1～5wt%程度であるのがさらに好ましい。結合樹脂の含有量が多過ぎると、磁気特性（特に最大磁気エネルギー積）の向上が図れず、また、寸法精度が低下する傾向を示す。また、結合樹脂の含有量が少な過ぎると、成形性が低下する。

【0055】3. 酸化防止剤

酸化防止剤は、混練物の製造の際等に、希土類磁石粉末の酸化劣化や結合樹脂の酸化による変質（希土類磁石粉末の金属成分が触媒として働くことにより生じる）を防止するために該組成物中に添加される添加剤である。この酸化防止剤の添加は、希土類磁石粉末の酸化を防止し、磁石の磁気特性の向上を図るのに寄与するとともに、希土類ボンド磁石用組成物の混練時、成形時における熱的安定性の向上に寄与し、少ない結合樹脂量で良好な成形性を確保する上で重要な役割を果たしている。

【0056】この酸化防止剤は、混練時や磁石への成形時等において揮発したり、変質したりするので、製造された希土類ボンド磁石中には、その一部が残留した状態で存在する。

【0057】酸化防止剤としては、希土類磁石粉末等の酸化を防止または抑制し得るものであればいかなるものでもよく、例えば、アミン系化合物、アミノ酸系化合物、ニトロカルボン酸類、ヒドラジン化合物、シアン化合物、硫化物等の、金属イオン、特にFe成分に対しキレート化合物を生成するキレート化剤が好適に使用される。なお、酸化防止剤の種類、組成等については、これらのものに限定されないことは言うまでもない。

【0058】このような酸化防止剤を添加する場合、混練物中の酸化防止剤の含有量は、0.1～2wt%程度とするのが好ましく、0.5～1.5wt%程度とするのがより好ましい。この場合、酸化防止剤の含有量は、結合樹脂の含有量に対し2～150%程度であるのが好ましく、30～100%程度であるのがより好ましい。

【0059】なお、本発明では、酸化防止剤の添加量は、前記範囲の下限值以下であってもよく、また、無添加であってもよいことは、言うまでもない。

【0060】前記結合樹脂と酸化防止剤との添加量は、次のようなことに留意して決定される。

【0061】すなわち、結合樹脂が少ないときには、相対的に磁石粉末量が増加して、混練の際の混練物の粘度が高くなり、混練トルクが増大し、発熱により樹脂の酸化が促進される傾向となる。この時、酸化防止剤量が少ないと、樹脂の酸化を十分に抑制することができなくな

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り、混練物（樹脂溶融物）の粘度上昇が生じて混練性、成形性が低下し、低空孔率、高機械的強度で寸法安定性に優れた磁石が得られない。また、酸化防止剤が多いと、相対的に樹脂量が減少し、成形体の機械的強度が低下する傾向を示す。

【0062】一方、結合樹脂が多いときには、相対的に磁石粉末量が減少して、磁石粉末の樹脂に対する影響が低下して樹脂の酸化が起こりにくくなる。そのため、酸化防止剤が少なくても樹脂の酸化を抑制することが可能となる。

【0063】このように、結合樹脂の含有量が比較的多ければ、酸化防止剤の含有量を少なくすることができ、逆に、結合樹脂の含有量が少なければ、酸化防止剤の含有量を多くする必要がある。

【0064】従って、混練物中の結合樹脂と酸化防止剤との合計含有量は、1.0～8.0wt%であるのが好ましく、2.0～6.0wt%であるのがより好ましい。このような範囲とすることにより、成形時における成形性、成形の容易性、磁石粉末等の酸化防止の向上に寄与し、低空孔率、高機械的強度、高磁気特性の磁石が得ら

れる。

【0065】4. その他の添加剤

また、混練物中には、必要に応じ、例えば、可塑剤（例えば、ステアリン酸亜鉛等の脂肪酸塩、脂肪酸）、潤滑剤（例えば、シリコンオイル、各種ワックス、脂肪酸、アルミナ、シリカ、チタニア等の各種無機潤滑剤）、その他成形助剤等の各種添加剤が添加されていて

もよい。

【0066】可塑剤の添加は、成形時の流動性を向上させるので、より少ない結合樹脂の添加量で同様の特性を得ることができ、また、より低い成形圧で圧縮成形することを可能とする。潤滑剤の添加についても同様である。可塑剤の添加量は、0.01～0.2wt%程度であるのが好ましく、潤滑剤の添加量は、0.05～0.5wt%程度であるのが好ましい。

【0067】以上のような希土類磁石粉末と、結合樹脂と、好ましくは酸化防止剤と、必要に応じその他の添加剤とを混合し、さらに混練して混練物を製造する。

【0068】混合は、例えば、ヘンシェルミキサー等の混合機や攪拌機を用いて行われる。

【0069】混練は、例えば2軸押出混練機、ロール式混練機、ニーダー等の混練機を用いて行われる。

【0070】この混練は、好ましくは用いる結合樹脂の熱変形温度（ASTM D648 による方法で測定）以上の温度、より好ましくは用いる結合樹脂の融点以上の温度で行われる。

【0071】例えば、結合樹脂としてポリアミド（熱変形温度145℃、融点178℃）を用いた場合には、好ましい混練温度は、150～280℃程度である。また、混練時間は、結合樹脂の種類や、混練温度等の諸条

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件により異なるが、通常は、5～40分程度とされる。

【0072】また、この混練は、希土類磁石粉末の表面が溶融または軟化した結合樹脂成分により覆われた状態となるように、十分に行われる。前記混練温度で混練した場合、このような状態を得るための混練時間は、結合樹脂の種類や使用する混練機、混練温度等の諸条件により異なるが、通常5～90分程度とするのが好ましく、5～60分程度とするのがより好ましい。

【0073】このような条件で混練することにより、混練の効率が向上し、常温で混練する場合に比べてより短時間で均一に混練することができるとともに、結合樹脂の粘度が下がった状態で混練されるので、希土類磁石粉末の周囲を結合樹脂が均一に覆った状態となり、混練物中の空孔率の減少、すなわち製造された磁石中の空孔率の減少に寄与する。

【0074】なお、結合樹脂として、n種類の熱可塑性樹脂を混合して用いる場合、前記「用いる結合樹脂の熱変形温度（または融点）」は、例えば、次のようにして換算することができる。

【0075】熱可塑性樹脂の合計を1重量部としたときの各熱可塑性樹脂の量をそれぞれ A_1 、 A_2 ・・・ A_n 重量部、各熱可塑性樹脂の熱変形温度（または融点）をそれぞれ T_1 、 T_2 ・・・ T_n としたとき、用いる熱可塑性樹脂の熱変形温度（または融点）は、 $A_1 T_1 + A_2 T_2 + \dots + A_n T_n$ で表される。なお、この換算は、以下の工程において、n種類の熱可塑性樹脂を混合して用いる場合にも同様とする。

【0076】＜2＞造粒物の製造

前記＜1＞で製造された混練物を造粒または整粒し、所定の粒径の粒状物を製造する。

【0077】造粒または整粒の方法は、特に限定されないが、混練物を粉砕することによりなされるのが好ましい。この粉砕は、例えば、ボールミル、振動ミル、破碎機、ジェットミル、ピンミル等を用いて行われる。

【0078】また、例えば押出式造粒機のような造粒機を用いて行うこともでき、さらには、造粒機による造粒と、前記粉砕とを組み合わせることもできる。

【0079】また、粒状物の粒径の調整は、篩い等を用いて分級することにより行うことができる。

【0080】粒状物の平均粒径は、10 μ m～2mm程度であるのが好ましく、20 μ m～2mm程度であるのがより好ましく、50 μ m～2mm程度であるのがさらに好ましい。粒状物の平均粒径が2mm以上では、特に成形される磁石の寸法が小さい場合に、すなわち成形金型のギャップの寸法が小さい場合に、粒状物の金型への充填量を微妙に調整することが困難となり、定量性が劣るので、ボンド磁石の寸法精度の向上が図れない。一方、平均粒径10 μ m以下の粒状物は、製造（造粒）が困難かまたは手間がかかる場合があり、また、平均粒径が小さ過ぎると、得られたボンド磁石の空孔率が上昇する傾向を示

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す。

【0081】このような粒状物は、粒径にある程度のバラツキがあるものでもよいが、粒径が均一なものが好ましい。これにより、金型への充填密度が増大し、低空孔率で寸法精度の高いボンド磁石が得られる。

【0082】なお、ここで言う粒状物は、粒径の大きいペレット（塊状物）とは区別される。

【0083】＜3＞加圧成形

前記＜2＞で得られた粒状物を用いて加圧成形を行う。以下、代表的な圧縮成形について説明する。

【0084】造粒物を圧縮成形機の金型内（ギャップ）に充填し、磁場中（配向磁場が例えば5～20kOe、配向方向は、縦、横、ラジアル方向のいずれも可）または無磁場中で圧縮成形する。

【0085】この圧縮成形は、温間成形で行われる。すなわち、成形金型を加熱する等により、成形時の材料温度を、用いる熱可塑性樹脂（結合樹脂）が軟化または熔融状態となる所定の温度（第1の温度）とする。

【0086】この第1の温度は、用いる熱可塑性樹脂の熱変形温度以上の温度とされる。さらには、用いる熱可塑性樹脂の融点以上の温度とされるのが好ましく、融点から（融点+200）℃程度までの範囲の所定の温度とされるのがより好ましく、融点から（融点+130）℃程度までの範囲の所定の温度とされるのがさらに好ましい。

【0087】例えば、用いる熱可塑性樹脂がポリアミド（融点：178℃）である場合、成形時における特に好ましい材料温度（第1の温度）は、180～300℃程度とされる。

【0088】このような温度で成形することにより、金型内での成形材料の流動性が向上し、円柱状、ブロック状のものは勿論のこと、円筒状（リング状）、平板状、湾曲板状等の薄肉部を有する形状のもの、小型のもの、長尺なものでも、低空孔率で、機械的強度が高く、良好かつ安定した形状、寸法のを量産することができる。

【0089】圧縮成形における成形圧力は、好ましくは60kgf/mm²以下、より好ましくは2～50kgf/mm²程度、さらに好ましくは5～40kgf/mm²程度とされる。本発明では、前述したような第1の温度で成形を行うため、このような比較的低い成形圧力でも、前述したような長所を持つボンド磁石を成形（賦形）することができる。

【0090】＜4＞冷却

加圧成形後、成形体を冷却する。この冷却は、少なくとも前記第1の温度未満である所定の温度（第2の温度）まで加圧状態で行う。以下、これを「加圧下冷却」と言う。

【0091】このような加圧下冷却を行うことにより、成形時の低空孔率な状態がそのまま維持されるので、低

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空孔率で寸法精度が高く、磁気特性に優れる希土類ボンド磁石が得られる。

【0092】第2の温度（除圧温度）は、得られたボンド磁石の空孔率の低減および寸法精度の向上にとって、できるだけ低い温度であるのが好ましく、本発明では、用いる熱可塑性樹脂の融点またはそれ以下の温度であるのが好ましく、用いる熱可塑性樹脂の熱変形温度（軟化点）またはそれ以下の温度であるのがより好ましい。

【0093】また、前記第1の温度と第2の温度との差は、20℃以上であるのが好ましく、50℃以上であるのがより好ましい。この温度差が大きい程、空孔率の低減および寸法精度の向上の効果が大きい。

【0094】なお、磁石粉末の含有量が比較的多い場合には、第2の温度をより高く設定しても低空孔率のボンド磁石を得易い。従って、例えば、混練物中の磁石粉末の含有量が例えば94wt%以上の場合には、第2の温度を、用いる熱可塑性樹脂の融点付近の温度または融点以上の温度（～融点+10℃程度）としても、空孔率を低く（4.5%以下または4.0%以下）することができる。

【0095】また、加圧下冷却は、加圧成形時の加圧を一旦解除または緩和した後、行ってもよいが、加圧成形時の加圧を解除することなく連続して行われるのが、工程の簡素化および寸法精度の向上等のために好ましい。

【0096】また、加圧下冷却の際の圧力は、一定でも変化してもよいが、少なくとも用いる熱可塑性樹脂の融点（特に熱変形温度）までは一定に保持されているのが好ましい。加圧下冷却の際の圧力が変化する場合、例えば、圧力が連続的または段階的に増加または減少するようなパターンを含んでいてもよい。

【0097】また、加圧下冷却の際の圧力（該圧力が経時変化する場合にはその平均圧力）は、加圧成形時の成形圧力と同等またはそれ以下であるのが好ましく、少なくとも用いる熱可塑性樹脂の融点までは加圧成形時の成形圧力と同等であるのがより好ましい。用いる熱可塑性樹脂の融点から熱変形温度までの間も加圧下で冷却する場合は、その間の圧力は、加圧成形時の成形圧力の40～100%程度とするのが好ましく、50～80%程度とするのがより好ましい。

【0098】なお、本発明では、加圧下冷却の後（除圧後）に、非加圧下（常圧下）で冷却を続行してもよいことは、言うまでもない。また、非加圧下冷却を行った後、再度加圧下冷却を行ってもよい。

【0099】加圧下冷却の際の冷却速度（冷却速度が経時変化する場合にはその平均値）は、特に限定されないが、0.5～100℃/秒であるのが好ましく、1～80℃/秒であるのがより好ましい。冷却速度が速過ぎると、冷却に伴う急速な収縮により、成形体内部に微細なクラックが発生し、機械的強度の低下を招くおそれがあり、また、冷却により内部応力が増大し、金型からの除

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材時に応力緩和によるひずみや変形が生じて、寸法精度が低下することがある。一方、冷却速度が遅過ぎると、成形のサイクルタイムが増加し、生産性が低下する。

【0100】また、除圧後にも冷却を続行する場合、その冷却速度は特に限定されず、前記と同様の冷却速度とすることができる。

【0101】なお、加圧下冷却の際および除圧後の冷却の際の冷却速度は、それぞれ、一定でも変化してもよい。

【0102】なお、本工程において、冷却の方法は、例えば自然空冷、強制空冷、水冷、油冷、水冷と空冷の組み合わせ等、いかなる方法を採用してもよい。

【0103】以上のような本発明の方法で製造された希土類ボンド磁石は、次のような優れた特性を有する。すなわち、空孔率が低く、好ましくは4.5% (vol%)以下、より好ましくは3.5%以下、さらに好ましくは2.0%以下とすることができる。このように、空孔率が低い(=密度が高い)ので、機械的強度が高く、耐食性に優れ、また、寸法精度が高く、量産した場合にも寸法のバラツキが少なく、寸法安定性に優れている。

【0104】さらに、磁気特性に優れており、特に、磁石粉末の組成、磁石粉末の含有量の多さ等から、等方性磁石であっても、優れた磁気特性を有する。

【0105】すなわち、無磁場中で成形された希土類ボンド磁石の場合、最大磁気エネルギー積(BH)maxが好ましくは6MG0e以上、より好ましくは8MG0e以上であり、磁場中で成形された希土類ボンド磁石の場合、最大磁気エネルギー積(BH)maxが12MG0e以上、より好ましくは13MG0e以上である。

【0106】なお、本発明により得られた希土類ボンド磁石の形状、寸法等は特に限定されず、例えば、形状に関しては、例えば、円柱状、角柱状、円筒状、円弧状(かわら状)、平板状、湾曲板状等のあらゆる形状のものが可能であり、その大きさも、大型のものから超小型のものまであらゆる大きさのものが可能である。

【0107】

【実施例】以下、本発明の具体的実施例について説明する。

【0108】(実施例1)下記の磁石粉末と結合樹脂(熱可塑性樹脂)と添加剤とを混合し、該混合物を混練し、該混練物を造粒(整粒)して粒状物を得、該粒状物を成形機の金型内に充填して無磁場中で圧縮成形(温間成形)し、成形時の加圧状態を同圧で維持しつつ冷却して、結合樹脂の固化により磁石粉末同士が結合された希土類ボンド磁石(サンプルNo. 1~9)を製造した。なお、各物質の含有量は、いずれも混練物中の量を示す。

【0109】・構成

Nd-Fe-B系磁石粉末: Nd 12.0 Fe 77.8 Co 4.3

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B5.9、96.0wt%

熱可塑性樹脂: 表1中に記載のA~G、各々2.8wt%

酸化防止剤: ヒドラジン系酸化防止剤、1.2wt%

混合: ヘンシェルミキサーを用いて混合。

【0110】混練: 2軸押出混練機により混練。混練温度は表2参照。

【0111】スクリー回転数100~300rpm。混練時間5~15分

造粒(整粒): 混練物を粉碎と分級により平均粒径0.8mmの粒に調整。

【0112】成形: 粒状物を金型に投入し、所定の成形温度(第1の温度)に加熱したところで加圧成形した。

【0113】成形温度、成形圧力は表2を参照。

【0114】冷却: 加圧状態を維持しつつ除圧温度(第2の温度)まで冷却し、除圧後さらに常温まで冷却して、サンプルを取り出した。

【0115】冷却方法は空冷とした。除圧温度は表2参照。

【0116】加圧下冷却での冷却速度は1℃/秒。

20 【0117】成形品形状: 円筒形状(外径φ30mm×内径φ28mm×高さ7mm)

平板形状(20mm角×厚さ3mm)(機械的強度測定用)
得られた希土類ボンド磁石について、磁気性能(磁束密度Br、保磁力iHc、最大磁気エネルギー積(BH)max)、密度、空孔率、機械的強度、耐食性を調べたところ、下記表3に示す通りであった。

【0118】なお、表3中の各測定項目の評価は、以下の方法に従った。

【0119】磁気性能: 40k0eでパルス着磁した後、最大印加磁場25k0eで直流磁気測定機により測定。または、成形サンプルから5mm角×厚さ1mmの磁石片を切り出した後、VSMで測定。

【0120】密度: アルキメデス法(水中法)により測定。

【0121】空孔率: 秤量組成と成形体の密度の測定値から算出。

【0122】機械的強度: 打ち抜きせん断試験により測定。試験機は(株)島津製作所製オートグラフを用い、円形ポンチ(外径3mm)により剪断速度1.0mm/minで行った。

【0123】試料には平板形状の磁石を使用。

【0124】耐食性: 温度80℃、湿度90%の恒温恒湿槽に、成形磁石を投入し、磁石表面に錆が発生するまでの時間を測定。表面観察は50時間毎に槽から取り出して光学顕微鏡(×10倍)で観察。500時間後は、500時間おきに観察を行った。

【0125】

【表1】

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樹脂 No.	結 合 樹 脂	融 点 [℃]	熱変形温度 [℃]
A	ポリアミド樹脂 (PA12)	178	145
B	ポリアミド樹脂 (コポリマPA6-12)	145	46
C	ポリアミド樹脂 (PA6)	215	180
D	ポリプロピレン樹脂 (PP)	174	105
E	ポリエチレン樹脂 (PE)	128	86
F	共重合ポリエステル	280	180
G	ポリフェニレンサルファイド (PPS)	287	260

【0126】

* * 【表2】

(実施例1)

サンプル No.	結 合 樹 脂	混練温度 [℃]	成形温度 [℃]	除圧温度 [℃]	成形圧力 [kgf/mm ²]
1	A	150~250	220	100	10
2	B	100~250	200	40	15
3	A (75%) + B (25%)	150~250	230	120	10
4	A (50%) + B (50%)	140~250	210	40	7.5
5	C	190~290	250	150	20
6	D	120~250	210	90	25
7	E	100~200	150	70	10
8	F	200~350	320	140	30
9	G	260~360	300	240	25

【0127】

※ ※ 【表3】

(実施例1)

サンプル No.	Br [kG]	iHc [kOe]	(BH) _{max} [MGoe]	密度 [g/cm ³]	空孔率 [%]	機械的強度 [kgf/mm ²]	耐食性 [時間]
1	7.21	9.26	10.1	6.01	0.59	7.90	>500
2	7.19	9.31	10.0	6.03	0.73	7.45	>500
3	7.23	9.23	10.1	6.03	0.37	7.78	>500
4	7.22	9.21	10.0	6.03	0.49	7.60	>500
5	7.27	9.27	10.0	6.08	0.92	8.10	300
6	7.24	9.35	10.3	5.88	0.58	6.95	300
7	7.23	9.30	10.2	5.95	0.38	5.80	350
8	7.03	9.12	9.8	6.29	0.64	9.65	450
9	7.01	9.10	9.8	6.27	0.59	9.73	450

【0128】表3から明らかなように、熱可塑性樹脂を結合樹脂として用いた本発明による希土類ボンド磁石(サンプルNo. 1~9)は、いずれも、低い成形圧力であるにもかかわらず、空孔率が1%以下と低く、ほぼ理論密度通りの高密度のボンド磁石が得られ、この結果、

非常に機械的強度の高い磁石を得ることができた。

【0129】また、磁石表面にコーティングを施さない状態でも、十分な耐食性を有していた。この理由は、空孔が少ないことにより、結合樹脂が磁石粉末表面を均一に覆っているためであると推定される。

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【0130】サンプルNo. 1～9の各磁石について、その切断面の電子顕微鏡写真（SEM）を撮影し、観察を行ったところ、空孔はほとんど観察されず、磁石粉末の周辺を結合樹脂成分が均一に分散しているのが確認された。

【0131】さらに、磁束密度Br、保磁力iHc、最大磁気エネルギー積(BH)maxが高く、優れた磁気特性であることがわかる。

【0132】（比較例1）下記の磁石粉末と結合樹脂（熱硬化性樹脂）とを混合し、該混合物を混練し、該混練物を造粒（整粒）して粒状物を得、該粒状物を成形機の金型内に充填して無磁場中で圧縮成形（冷間成形または温間成形）し、その後、結合樹脂を硬化させて、希土類ボンド磁石（サンプルNo. 10～15）を製造した。なお、各物質の含有量は、いずれも混練物中の量を示す。

【0133】・構成

Nd-Fe-B系磁石粉末：Nd12.0Fe77.8Co4.3B5.9、96.0wt%

熱硬化性樹脂：表4中に記載のもの、4.0wt%（硬化剤を含む）

混合：室温で固体の樹脂を用いた場合は、V型混合機で混合。

【0134】室温で液状の樹脂を用いた場合は、攪拌機で混合。

【0135】混練：ニーダーを用いて混練。混練温度は表5を参照。

*

樹脂 No.	結 合 樹 脂	軟化温度 [℃]	硬化条件
H	ビスフェノールA型エポキシ樹脂	室温以下	150℃×1hr
I	フェノールノボラック型エポキシ樹脂	80	170℃×2hr
J	フェノール樹脂	70	180℃×4hr

【0147】

※ ※【表5】

(比較例1)

サンプル No.	結 合 樹 脂	混 練 温 度 [℃]	成 形 温 度 [℃]	除 圧 温 度 [℃]	成 形 圧 力 [kgf/mm ²]
10	H	室温	室温	室温	20
11	H	室温	室温	室温	70
12	I	80～100	120	50	20
13	I	80～100	120	50	70
14	J	70～90	100	50	20
15	J	70～90	100	50	70

【0148】

【表6】

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*【0136】ニーダー回転数50～250rpm。混練時間30分。

【0137】造粒（整粒）：混練物を粉碎と分級により平均粒径0.8mm以下の粒に調整。

【0138】成形：粒状物を金型に投入し、所定の成形温度で加圧成形した。

【0139】成形温度、成形圧力は表5を参照。

【0140】冷却：除圧温度まで冷却し（サンプルNo. 10、11を除く）、除圧後さらに常温まで冷却して、サンプルを取り出した。

【0141】冷却方法は空冷とした。除圧温度は表5参照。

【0142】冷却速度は2℃/秒。

【0143】熱処理：仮成形品を恒温槽に入れ、熱硬化性樹脂の硬化を行う。

【0144】硬化条件は表4を参照。

【0145】成形品形状：円筒形状（外径φ30mm×内径φ28mm×高さ7mm）

平板形状（20mm角×厚さ3mm）（機械的強度測定用）
得られた希土類ボンド磁石について、磁気性能（最大磁気エネルギー積(BH)max）、密度、空孔率、機械的強度、耐食性を調べたところ、下記表6に示す通りであった。なお、各項目の評価方法は、実施例1と同様である。

【0146】

【表4】

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(比較例1)

サンプル No.	(BH) max [MGOe]	密度 [g/cm ³]	空孔率 [%]	機械的強度 [kgf/mm ²]	耐食性 [時間]
10	8.2	5.70	7.7	測定不能	50
11	9.1	5.85	5.27	3.86	200
12	8.0	5.67	8.18	測定不能	50
13	9.0	5.82	5.75	3.91	200
14	8.2	5.73	9.51	測定不能	50
15	9.2	5.90	6.83	4.01	150

【0149】表6から明らかなように、結合樹脂として熱硬化性樹脂を用いた比較例の磁石（サンプルNo. 10～15）では、成形圧力を20kgf/mm²としたときは勿論のこと、成形圧力を70kgf/mm²としたときでも、空孔率が高く、磁石成形体の密度が低い。この結果、磁石の機械的強度が低く、また耐食性も低い。

【0150】サンプルNo. 10～15の各磁石について、その切断面の電子顕微鏡写真（SEM）を撮影し、観察を行ったところ、内部に空孔が多く存在していた。また、その空孔の分布は、加圧成形時の圧力伝達の関係で、中心部に空孔が多く、表面付近に少ないというように不均一な状態となっていた。また、樹脂成分が偏析しているのが観察された。

【0151】また、成形圧力を20kgf/mm²としたときには、サンプル内での機械的強度のバラツキが大きく、そのため円形ポンチによって荷重をかけた場所以外のところで割れやクラックを生じたため、正確な機械的強度の測定ができなかった。一方、成形圧力を70kgf/mm²としたときには、混練物中の樹脂成分が漏れ、これによるバリが発生した。

【0152】（実施例2）下記の磁石粉末と結合樹脂（熱可塑性樹脂）と添加剤とを混合し、該混合物を混練し、該混練物を造粒（整粒）して粒状物を得、該粒状物を成形機の金型内に充填して磁場中で圧縮成形（温間成形）し、成形時の加圧状態を同圧で維持しつつ冷却して、希土類ボンド磁石（サンプルNo. 16～19）を製造した。なお、各物質の含有量は、いずれも混練物中の量を示す。

【0153】・構成

Sm-Co系磁石粉末：Sm（Co_{bal.} Fe 0.32 Cu 0.06 Zr 0.016）7.8、95.0wt%

熱可塑性樹脂：PPS樹脂、4.2wt%

酸化防止剤：ヒドラジン系酸化防止剤、0.8wt%
混合：V型混合機を用いて混合。

20 【0154】混練：各種混練機を使用。混練条件は表7を参照。

【0155】造粒（整粒）：混練物を粉砕と分級により平均粒径0.8mmの粒に調整。

【0156】成形：粒状物を金型に投入し、所定の成形温度（第1の温度）に加熱したところで、横磁場（15kOe）を印加しながら加圧成形した。

【0157】成形温度は320℃、成形圧力は20kgf/mm²とした。

30 【0158】冷却：加圧状態を維持しつつ除圧温度（第2の温度）150℃まで冷却し、除圧後さらに常温まで冷却して、サンプルを取り出した。

【0159】冷却方法は空冷とした。

【0160】加圧下冷却での冷却速度は5℃/秒。

【0161】成形品形状：直方体（縦11mm×横8mm×高さ7mm、高さ方向が配向方向。）

平板形状（20mm角×厚さ3mm）（機械的強度測定用）
得られた希土類ボンド磁石について、磁気性能（最大磁気エネルギー積(BH)max）、密度、空孔率、機械的強度、耐食性を調べたところ、下記表8に示す通りであった。なお、各項目の評価方法は、実施例1と同様である。

【0162】

【表7】

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(実施例2)

サンプル No.	混練機	混練温度 [℃]	回転数 [rpm]	処理量
16	二軸押出混練機	170~320	100~250	30kg/hr
17	ロール混練機	180~300	10~100	5kg/batch, 15min/batch
18	ニーダー	180~300	20~100	10kg/batch, 30min/batch
19	KCK	170~320	20~80	20kg/hr

【0163】

* * 【表8】

(実施例2)

サンプル No.	(BH) _{max} [MGDe]	密度 [g/cm ³]	空孔率 [%]	機械的強度 [kgf/mm ²]	耐食性 [時間]
16	15.2	6.63	0.65	8.14	>1000
17	15.5	6.65	0.35	8.23	>1000
18	14.9	6.61	0.95	8.09	>1000
19	15.3	6.63	0.65	8.19	>1000

【0164】表8から明らかなように、本発明による希土類ボンド磁石（サンプルNo. 16~19）は、いずれも、空孔率が1%以下と低く、高密度のボンド磁石が得られ、この結果、機械的強度および耐食性が高いものであった。

【0165】また、サンプルNo. 16~19の各磁石について、前記と同様に電子顕微鏡写真（SEM）を撮影し、観察を行ったところ、空孔はほとんど観察されず、磁石粉末の周辺を結合樹脂成分が均一に分散しているのが確認された。

【0166】さらに、最大磁気エネルギー積 (BH)_{max} が高く、優れた磁気特性であることがわかる。

【0167】（比較例2）下記の磁石粉末と結合樹脂（熱可塑性樹脂）と添加剤とを混合し、該混合物を成形機の金型内に充填して磁場中で圧縮成形（温間成形）し、希土類ボンド磁石（サンプルNo. 20、21）を製造した。なお、各物質の含有量は、いずれも混合物中の量を示す。

【0168】・構成

Sm-Co系磁石粉末：Sm (Co_{bal}. Fe_{0.32} Cu_{0.06} Zr_{0.016}) 7.8、95.0wt%（サンプルNo. 20）、96.0wt%（サンプルNo. 21）

熱可塑性樹脂：PPS樹脂、4.2wt%（サンプルNo. 20）、3.2wt%（サンプルNo. 21）

酸化防止剤：ヒドラジン系酸化防止剤、0.8wt%
混合：V型混合機を用いて混合。

【0169】成形：混合物を金型に投入し、所定の成形

温度に加熱したところで、横磁場（15kOe）を印加しながら加圧成形した。

【0170】成形温度は320℃、成形圧力は20kgf/mm²とした。

【0171】冷却：温度150℃まで冷却し、サンプルを取り出した。

【0172】冷却方法は空冷とした。

【0173】冷却速度は5℃/秒。

【0174】成形品形状：直方体（縦11mm×横8mm×高さ7mm、高さ方向が配向方向。）

平板形状（20mm角×厚さ3mm）（機械的強度測定用）
サンプルNo. 20および21の磁石は、いずれも、成形時に樹脂漏れが生じ、成形品のエッジや端面部が成形機のパンチに付着することにより、成形品のえぐれが生じたり、エッジ等のかけが生じて、所望の形状を得ることができなかった。

【0175】成形された部分の電子顕微鏡写真（SEM）を撮影し、観察を行ったところ、結合樹脂成分の分散は不均一となっており、磁石粉末部分と結合樹脂部分が混在した状態であった。

【0176】また、上述したように、サンプルNo. 20および21の磁石は、いずれも、不良品であったため、機械的強度等の有効な測定はできなかった。

【0177】（実施例3）下記の磁石粉末（2種）と結合樹脂（熱可塑性樹脂）と添加剤とを混合し、該混合物を混練し、該混練物を造粒（整粒）して粒状物を得、該粒状物を成形機の金型内に充填して磁場中で圧縮成形

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(温間成形)し、成形時の加圧状態を同圧で維持しつつ冷却して、希土類ボンド磁石(サンプルNo. 22~30)を製造した。なお、各物質の含有量は、いずれも混練物中の量を示す。

【0178】・構成

Sm-Co系磁石粉末: Sm (Co 0.672 Fe 0.22 Cu 0.08 Zr 0.028) 8.35、70.5wt%

Sm-Fe-N系磁石粉末: Sm₂ Fe₁₇ N₃、23.5wt%

熱可塑性樹脂: ポリアミド樹脂(ナイロン12)、5.0wt%

酸化防止剤: フェノール系酸化防止剤、1.0wt%

混合: ヘンシェルミキサーを用いて混合。

【0179】混練: 2軸押出混練機により混練。混練温度は150~300℃。

【0180】スクリー回転数100~300rpm。混練時間10分

造粒(整粒): 混練物を粉碎と分級により表9に示す粒*

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* 度に調整。

【0181】成形: 粒状物をすり切り方式で金型に投入し、220℃(第1の温度)に加熱したところで、横磁場(15kOe)を印加しながら加圧成形した。成形圧力は10kgf/mm²とした。

【0182】冷却: 加圧状態を維持しつつ除圧温度(第2の温度)100℃まで冷却し、サンプルを取り出した。

【0183】冷却方法は水冷とした。

【0184】加圧下冷却での冷却速度は20℃/秒。

【0185】成形品形状: 平板形状(幅15mm×高さ2.5mm×高さ5mm、高さ方向が配向方向)

得られた希土類ボンド磁石について、磁石の重量、密度、空孔率、高さを測定したところ、下記表9に示す通りであった。

【0186】

【表9】

(実施例3)

サンプル No.	粒状物の平均 粒径 [mm]	成形品重量 [mg]	密度 [g/cm ³]	空孔率 [%]	高さ [mm]
22	2	1073	5.78	0.30	4.95
23	1.8	1075	5.78	0.30	4.96
24	1.5	1077	5.78	0.30	4.97
25	1	1079	5.78	0.30	4.98
26	0.5	1083	5.79	0.12	4.99
27	0.1	1081	5.79	0.12	4.98
28	0.05	1080	5.76	0.64	5.00
29	0.01	1075	5.72	1.33	5.01
30	0.007	1071	5.68	2.02	5.03

注) 成形品重量と高さは、n=10の平均値

【0187】表9から明らかなように、粒状物の粒径の設定により、優れた定量性が得られ、低空孔率でかつ寸法精度の高いボンド磁石が得られる。特に、粒状物の粒径が0.01~2mmの範囲である場合には、超低空孔率(1%以下)と、高い寸法精度(寸法誤差が±5/100mm以内)と両立することができた。

【0188】(実施例4、比較例3)下記の磁石粉末と結合樹脂(熱可塑性樹脂)と添加剤とを混合し、該混合物を混練し、該混練物を造粒(整粒)して粒状物を得、該粒状物を成形機の金型内に充填して磁場中で圧縮成形(温間成形)し、成形時の加圧状態を同圧で維持しつつ冷却して、希土類ボンド磁石(サンプルNo. 31~42)を製造した。なお、各物質の含有量は、いずれも混練物中の量を示す。

【0189】・構成

Nd-Fe-B系磁石粉末: Nd_{12.6} Fe_{69.3} Co_{12.0}

B_{6.0} Zr_{0.1}、97.0wt%

熱可塑性樹脂: 表1中のAまたはF、各々1.5wt%

酸化防止剤: ヒドラジン系酸化防止剤、1.4wt%

潤滑剤: ステアリン酸亜鉛、0.1wt%

混合: ヘンシェルミキサーを用いて混合。

【0190】混練: 2軸押出混練機により混練。混練温度は150~350℃。

【0191】スクリー回転数100~300rpm。混練時間5分

造粒(整粒): 混練物を粉碎と分級により平均粒径0.3mmの粒に調整。

【0192】成形: 粒状物を金型に投入し、表10に示す成形温度(第1の温度)に加熱したところで、ラジアル磁場(15kOe)を印加しながら加圧成形した。成形圧力は15kgf/mm²とした。

【0193】冷却: 加圧状態を維持しつつ除圧温度(第

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2の温度) 100℃まで冷却し、除圧後さらに常温まで冷却して、サンプルを取り出した。

【0194】冷却方法は水冷とした。

【0195】加圧下冷却での冷却速度は30℃/秒。

【0196】成形品形状：円筒形状（外径φ20mm×内径φ18mm×高さ5mm、高さ方向に加圧）

平板形状（20mm角×厚さ3mm）（機械的強度測定用）

得られた希土類ボンド磁石（実施例4：サンプルNo. 3*

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* 2～36、38～42、比較例3：サンプルNo. 31、37）について、磁気性能（最大磁気エネルギー積(BH)_{max}）、密度、空孔率、機械的強度を調べたところ、下記表10に示す通りであった。なお、各項目の評価方法は、実施例1と同様である。

【0197】

【表10】

（実施例4、比較例3）

サンプル No.	結合樹脂	成形温度 [℃]	(BH) _{max} [MG0e]	密度 [g/cm ³]	空孔率 [%]	機械的強度 [kgf/mm ²]
31	A	130	測定不能	測定不能	測定不能	測定不能
32	A	150	17.0	6.21	2.52	5.10
33	A	180	17.5	6.28	1.42	7.10
34	A	200	18.2	6.34	0.48	7.70
35	A	300	17.8	6.32	0.79	7.61
36	A	360	16.2	6.32	0.79	7.55
37	F	160	測定不能	測定不能	測定不能	測定不能
38	F	190	16.9	6.36	2.70	8.10
39	F	250	17.5	6.44	1.48	9.25
40	F	300	18.0	6.50	0.56	9.78
41	F	350	17.6	6.50	0.56	9.65
42	F	400	15.5	6.50	0.56	9.60

【0198】表10中のサンプルNo. 32～36、38～42（実施例4）のように、成形温度が結合樹脂の熱変形温度以上のときには、成形時に結合樹脂が軟化または熔融状態となり、成形が可能であった。

【0199】特に、サンプルNo. 33～36、40、42のように、成形温度が結合樹脂の融点以上のときには、得られた磁石の空孔率がさらに低減し、磁気性能もより高くなる。

【0200】これに対し、サンプルNo. 31、37（比較例3）のように、成形温度が結合樹脂の熱変形温度未満のときには、成形時に結合樹脂が軟化しないため、粒状物が互いに固着せず、そのため、形状を保持することができず、成形不能または成形不良であった。従って、各測定項目についても、測定不能であった。

【0201】（実施例5、比較例4）下記の磁石粉末と結合樹脂（熱可塑性樹脂）と添加剤とを混合し、該混合物を混練し、該混練物を造粒（整粒）して粒状物を得、該粒状物を成形機の金型内に充填して無磁場中で圧縮成形（温間成形）し、成形時の加圧状態を同圧で維持しつつ冷却して、希土類ボンド磁石（サンプルNo. 43～52）を製造した。なお、各物質の含有量は、いずれも混練物中の量を示す。

【0202】・構成

ナノ結晶Nd-Fe-B系磁石粉末：Nd_{5.5}Fe₆₆B 50

18.5Co₅Cr₅、98.0wt%

熱可塑性樹脂：表1中のAまたはG、各々1.0wt%

酸化防止剤：ヒドラジン系酸化防止剤 1.0wt%

30 混合：ヘンシェルミキサーを用いて混合。

【0203】混練：2軸押出混練機により混練。混練温度は150～350℃。

【0204】スクリー回転数100～300rpm。混練時間10分

造粒（整粒）：混練物を粉碎と分級により平均粒径0.1mmの粒に調整。

【0205】成形：粒状物を金型に投入し、所定の成形温度（第1の温度）に加熱したところで、加圧成形した。成形温度は、200℃（樹脂A）および300℃（樹脂G）、成形圧力は25kgf/mm²とした。

40 【0206】冷却：加圧状態を維持しつつ、表11に示す除圧温度（第2の温度）まで冷却し、サンプルを取り出した。冷却方法は水冷とした。

【0207】加圧下冷却での冷却速度は50℃/秒。

【0208】成形品形状：円筒形状（外径φ10mm×内径φ7mm×高さ7mm、高さ方向に加圧）

得られた希土類ボンド磁石（実施例5：サンプルNo. 44～47、49～52、比較例4：サンプルNo. 43、48）について、磁気性能（最大磁気エネルギー積(BH)_{max}）、密度、空孔率、外径を調べたところ、下記表1

(15)

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1に示す通りであった。なお、各項目の評価方法は、実施例1と同様である。

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*【0209】

*【表11】

(実施例5、比較例4)

サンプル No.	結合樹脂	除圧温度 [℃]	(BH) max [MG0e]	密度 [g/cm ³]	空孔率 [%]	外径 [mm]
43	A	200	8.0	6.49	3.55	10.00±0.08
44	A	180	8.2	6.56	2.51	10.00±0.05
45	A	160	8.6	6.65	1.17	10.00±0.02
46	A	140	8.6	6.67	0.87	10.01±0.02
47	A	100	8.7	6.68	0.73	10.01±0.01
48	G	300	7.3	6.55	4.26	10.04±0.10
49	G	275	7.9	6.64	2.95	10.02±0.04
50	G	260	8.3	6.74	1.49	10.01±0.03
51	G	240	8.5	6.78	0.90	10.00±0.01
52	G	200	8.5	6.79	0.76	10.00±0.01

注) 外径は、n=10の測定値から算出

【0210】表11中のサンプルNo. 44～47、49～52（実施例5）のように、除圧温度が結合樹脂の融点以下または除圧温度と成形温度との差が20℃以上のときには、得られた磁石の空孔率が低く、密度が高く、磁気性能が高く、寸法精度が高い（寸法誤差が±5/100mm以内）。このような特性は、除圧温度が低いほど、向上している。

【0211】特に、サンプルNo. 46、47、50、51のように、除圧温度が結合樹脂の熱変形温度以下ときには、ほぼ理論密度に近い密度を達成することができ、磁石粉末の特性を十分に発揮させることが可能な極めて優れた磁気性能の磁石となる。

【0212】これに対し、サンプルNo. 43、48（比較例4）のように、除圧温度と成形温度が同一である場合、寸法精度が低く、空孔率も前記サンプルNo. 44～47、49～52に比べて高い。

【0213】

【発明の効果】以上述べたように、本発明によれば、混練物の粒状物を用いて温間成形により加圧成形を行うこ

と、さらには、温間成形の後、所定温度まで加圧状態で冷却を行うことにより、少ない結合樹脂量でも成形性に優れ、低空孔率で、機械的強度が高く、また、寸法安定性（寸法精度）が高く、磁気特性に優れた希土類ボンド磁石を提供することができる。

【0214】この場合、粒状体の粒径が所望の範囲である場合には、空孔率が極めて低く、しかも、寸法安定性がさらに向上する。

【0215】また、熱可塑性樹脂を軟化または熔融状態として加圧成形するので、比較的低い成形圧力で上記特性の希土類ボンド磁石を製造することができ、製造が容易である。

【0216】特に、冷却時における第2の温度（除圧温度）が、用いる熱可塑性樹脂の融点以下の温度、さらには熱変形温度以下の温度である場合や、第1の温度と所定温度以上乖離している場合には、空孔率が極めて低くかつ寸法安定性が極めて高い希土類ボンド磁石を提供することができる。